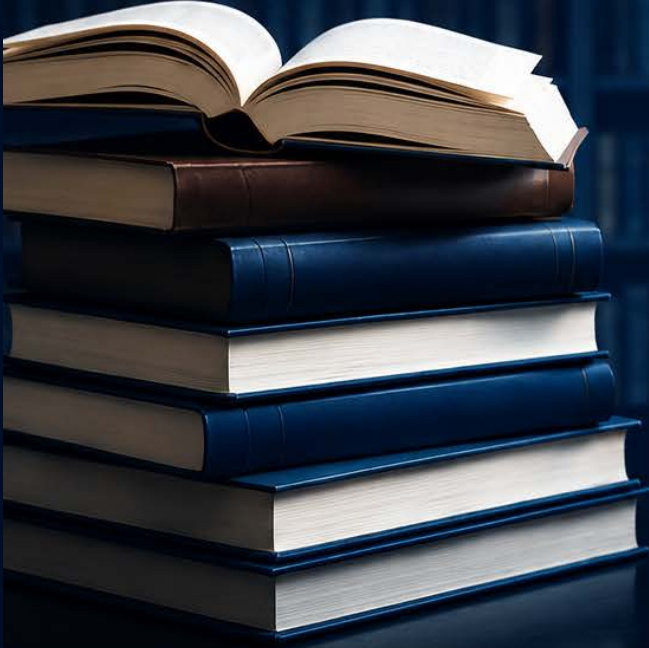


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Editör: Dr.Öğr.Üyesi Murat Metehan TÜRKOĞLU



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Güncel Konular ve Akademik Değerlendirmeler

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Dr.Öğr.Üyesi Murat Metehan TÜRKOĞLU

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İÇİNDEKİLER

Model-Based Evaluation of Medical ^{89}Zr Production Via The $^{89}\text{Y}(\text{p},\text{n})^{89}\text{Zr}$ and $^{89}\text{Y}(\text{d},2\text{n})^{89}\text{Zr}$ Reactions Using Talys	1
<i>Murat DAĞ</i>	
Comparative Talys Analysis of Medical ^{120}I Production Via The $^{120}\text{Te}(\text{p},\text{n})$ and $^{122}\text{Te}(\text{p},3\text{n})$ Reactions	19
<i>Murat DAĞ</i>	
BaO Katkılı PLA Kompozitlerinin Radyasyon Zırhlama Özelliklerinin XCOM ve Phy-X/PSD ile İncelenmesi	37
<i>Seda ERDÖNMEZ</i>	
Boron-Hydride Systems: Superconductivity Under High Pressure and the Quest for Room-Temperature Superconductivity	58
<i>Özge ERDEM</i>	
Topraktan Sofraya Doğal Radyonüklitler: Gıda Zincirinde Transfer, İç Doz ve Radyolojik Risk.....	73
<i>Gülçin Bilgici CENGİZ</i>	
The Relationship Between Radon Gas Emission and Earthquakes.....	95
<i>Selcen UZUN DURAN</i>	

"Bu kitapta yer alan bölümlerde kullanılan kaynakların, görüşlerin, bulguların, sonuçların, tablo, şekil, resim ve her türlü içeriğin sorumluluğu yazar veya yazarlarına ait olup ulusal ve uluslararası telif haklarına konu olabilecek mali ve hukuki sorumluluk da yazarlara aittir."

MODEL-BASED EVALUATION OF MEDICAL 89ZR PRODUCTION VIA THE 89Y(p,n)89ZR AND 89Y(d,2n)89ZR REACTIONS USING TALYS

Murat DAĞ¹

1. INTRODUCTION

Zirconium-89 (89Zr) has emerged as a key positron-emitting radionuclide for immuno-PET, particularly because its physical half-life matches the biological kinetics of monoclonal antibodies. Its relatively long half-life (78.41 h) enables efficient radiolabeling of monoclonal antibodies and other slowly circulating biomolecules, thereby allowing high-quality PET imaging several days after radiopharmaceutical administration. This characteristic has established 89Zr as an indispensable radionuclide for cancer diagnosis, pharmacokinetic investigations, personalized medicine, and theranostic applications (Deri et al., 2013; Holland et al., 2009). Accordingly, substantial research has focused on developing and optimizing production routes that simultaneously achieve high radionuclidic purity and satisfactory production yields.

Cyclotron production of 89Zr is commonly performed using monoisotopic 89Y targets. Because natural yttrium consists almost entirely of 89Y, target preparation and reaction analysis are considerably simplified compared with multi-isotopic target materials. Among the available charged-particle-induced reactions, the 89Y(p,n)89Zr reaction is the most widely used

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production route owing to its relatively low threshold energy and compatibility with conventional medical cyclotrons. Numerous experimental investigations have demonstrated that this reaction provides high production cross sections while maintaining acceptable radionuclidic purity when an appropriate proton-energy window is selected (Johnson et al., 1968; Uddin et al., 2005; Omara et al., 2009). In contrast, the $^{89}\text{Y}(d,2n)^{89}\text{Zr}$ reaction has been proposed as an alternative production route capable of providing comparable or even higher production yields under suitable irradiation conditions (Bissem et al., 1980; Uddin et al., 2007; Lebeda et al., 2015). However, deuteron-induced reactions involve more complicated reaction mechanisms because of deuteron breakup, stripping reactions, pre-equilibrium emission, and compound-nucleus formation, making theoretical prediction more challenging than for proton-induced reactions.

Reliable prediction of excitation functions is essential for optimizing irradiation conditions, estimating The TALYS codeproduction performance, and minimizing radionuclidic impurities during medical radionuclide production. Modern nuclear reaction codes provide an efficient theoretical framework for performing such investigations before experimental production. Among these codes, TALYS has become one of the most widely used tools for calculating excitation functions, reaction cross sections, radionuclide activities, and production yields over a broad range of incident particles and target nuclei (Koning et al., 2023; Koning & Rochman, 2012). Nevertheless, the accuracy of TALYS calculations depends strongly on the adopted nuclear models, particularly the nuclear level-density model governing the statistical decay of the compound nucleus.

Nuclear level density is one of the most influential quantities in statistical nuclear reaction calculations because it governs particle-emission probabilities and therefore has a direct influence on the calculated excitation functions. Among its

phenomenological formulations, the Constant Temperature Model (CTM), the Back-Shifted Fermi Gas Model (BSFGM), and the Generalized Superfluid Model (GSM) are the approaches most commonly employed for charged-particle-induced reaction calculations (Gilbert & Cameron, 2011; Ignatyuk et al., 1979). Although these models have been successfully applied to numerous nuclear reactions, their predictive performance may vary depending on the target nucleus, projectile, and reaction channel. Therefore, systematic comparison of these models using experimental excitation-function data remains important for improving the reliability of theoretical production studies.

Another important aspect of theoretical and experimental ^{89}Zr production studies is the definition of the reported reaction product. TALYS calculates the ground (^{89}gZr) and metastable (^{89}mZr) states independently, whereas some experimental datasets reported in the literature correspond to cumulative production after the isomeric transition of the short-lived metastable state (Satheesh et al., 2012). Therefore, in addition to the ground-state excitation functions, total and cumulative excitation functions were also included in the excitation-function analysis to enable physically meaningful comparisons with the available experimental datasets according to their respective measurement definitions.

Although numerous experimental and theoretical studies have investigated the two production pathways, most have primarily focused on reproducing excitation functions or reporting production yields (Mustafa et al., 1988; Khandaker et al., 2012; Manenti et al., 2019). Comparatively fewer studies have systematically assessed the predictive performance of different level-density models using quantitative statistical criteria while simultaneously considering both ground, total, and cumulative excitation functions. Such an evaluation is particularly valuable because visual agreement alone may not always identify the

model that most accurately reproduces the available experimental data.

In the present study, the two medical production routes were investigated using the TALYS nuclear reaction code with three phenomenological nuclear level-density models: the CTM, the BSFGM, and the GSM. Ground-state, total, and cumulative excitation functions were calculated and compared with the available experimental datasets. The predictive performance of each level-density model was quantitatively evaluated using the root mean square error (RMSE), chi-square (χ^2), and chi-square per experimental point (χ^2/N). Finally, the statistically best-performing level-density model for each reaction was employed in the TALYS medical isotope production module to calculate the total initial production yield (GBq/mAh) as a function of the incident particle energy for both proton- and deuteron-induced production routes. This combined evaluation provides a consistent theoretical framework for comparing the two production pathways and for identifying the most suitable modelling approach for medical ^{89}Zr production.

2. METHODS

The theoretical production of ^{89}Zr through the $^{89}\text{Y}(p,n)^{89}\text{Zr}$ and $^{89}\text{Y}(d,2n)^{89}\text{Zr}$ reactions was investigated using the TALYS nuclear reaction code. These reactions were selected because they represent the principal proton- and deuteron-induced production routes for medical ^{89}Zr and provide an appropriate basis for comparing different reaction mechanisms under identical theoretical conditions (Omara et al., 2009; Lebeda et al., 2015). The $^{89}\text{Y}(p,n)$ reaction proceeds predominantly through single-neutron emission, whereas the $^{89}\text{Y}(d,2n)$ reaction involves two-neutron emission together with more complex reaction mechanisms associated with deuteron interactions.

The TALYS nuclear reaction code was used as the computational tool for modelling nuclear reactions induced by neutral and charged particles in target nuclei with mass numbers greater than or equal to 12. The code combines optical-model calculations, direct reactions, pre-equilibrium emission, compound-nucleus formation, and statistical decay within a unified framework, allowing the calculation of excitation functions, residual radionuclide production cross sections, activities, and production yields (Koning et al., 2023; Koning & Rochman, 2012).

All calculations were performed using identical calculation conditions for the investigated reactions. The projectile energy was varied over the energy region relevant to medical cyclotron production, while the nuclear level-density model was treated as the principal variable in order to evaluate its influence on the calculated excitation functions.

The statistical decay of the compound nucleus depends strongly on the nuclear level density, which represents the number of available nuclear states at a given excitation energy. In TALYS, the total level density is obtained by summing the contributions from all spin and parity states,

$$\rho_{\text{tot}}(E_x) = \sum_{J, \Pi} \rho(E_x, J, \Pi) \quad (1)$$

A commonly adopted analytical description of the level density is provided by the Fermi-gas model (Gilbert & Cameron, 2011),

$$\rho_F^{\text{tot}}(E_x) = \frac{\sqrt{\pi} \exp(2\sqrt{aU})}{12\sqrt{2} a^{1/4} U^{5/4} \sigma} = \frac{\omega_F^{\text{tot}}(E_x)}{\sqrt{2\pi}\sigma} \quad (2)$$

where U denotes the effective excitation energy, σ is the spin cut-off parameter, and a is the level-density parameter. To

account for shell effects, the excitation-energy-dependent level-density parameter is expressed as

$$a(E_x) = \tilde{a} \left[1 + \delta W \frac{1 - \exp(-\gamma U)}{U} \right] \quad (3)$$

where the asymptotic level-density parameter is

$$\tilde{a} = \alpha A + \beta A^{2/3} \quad (4)$$

In these expressions, δW is the shell-correction energy, γ is the shell-damping parameter, and α and β are global constants (Ignatyuk et al., 1979).

Three phenomenological level-density models implemented in TALYS were investigated in this study: CTM assumes a constant nuclear temperature at low excitation energies and gradually merges with the Fermi-gas description at higher excitation energies. BSFGM employs the Fermi-gas formalism while accounting for pairing effects through an empirical back-shift parameter. GSM incorporates pairing correlations based on the superfluid model and describes the transition from the superfluid regime at low excitation energies to the normal Fermi-gas region at higher excitation energies.

These models were selected because they are among the most widely used phenomenological level-density approaches available in TALYS and have been extensively applied in charged-particle-induced reaction calculations (Koning et al., 2023).

TALYS calculates the production cross sections of the ground (^{89}gZr) and metastable (^{89}mZr) states independently. According to the decay data available in the National Nuclear Data Center (NNDC), ^{89}mZr decays to the ground state through isomeric transition with a branching ratio of 93.77%. Therefore, cumulative excitation functions were additionally calculated to

enable direct comparison with cumulative experimental datasets. The cumulative production cross section was obtained using

$$\sigma_{\text{cum}}(E) = \sigma_g(E) + b_{\text{IT}} \sigma_m(E) \quad (5)$$

where $\sigma_g(E)$ and $\sigma_m(E)$ denote the independently calculated ground-state and metastable-state production cross sections, respectively, and $b_{\text{IT}} = 0.9377$ is the isomeric-transition branching ratio of ^{89}mZr adopted from the NNDC decay data (National Nuclear Data Center, 2025).

Consequently, independent theoretical calculations were compared with independent experimental datasets, whereas cumulative theoretical calculations were compared with cumulative experimental measurements whenever such data were available.

Experimental excitation-function data for both production routes were compiled from the EXFOR database together with the corresponding original publications (Otuka et al., 2014). The available datasets were used to evaluate the predictive capability of the investigated level-density models over the entire energy range considered.

For each experimental data point, the theoretical cross section was obtained at the corresponding proton energy by interpolation of the calculated excitation function. The predictive performance of each level-density model was evaluated using the root mean square error (RMSE), chi-square (χ^2), and chi-square per experimental point (χ^2/N). The RMSE was calculated as

$$\text{RMSE} = \sqrt{\frac{1}{N} \sum_{i=1}^N (\sigma_i^{\text{calc}} - \sigma_i^{\text{exp}})^2} \quad (6)$$

where N is the total number of experimental data points, and σ_{calc} and σ_{exp} are the calculated and experimental cross sections, respectively. The chi-square value was calculated as

$$\chi^2 = \sum_{i=1}^N \left[\frac{\sigma_i^{\text{calc}} - \sigma_i^{\text{exp}}}{\Delta \sigma_i^{\text{exp}}} \right]^2 \quad (7)$$

where $\Delta \sigma_{\text{exp}}$ denotes the experimental cross-section uncertainty. The chi-square value per experimental point was then obtained from

$$\frac{\chi^2}{N} = \frac{1}{N} \sum_{i=1}^N \left[\frac{\sigma_i^{\text{calc}} - \sigma_i^{\text{exp}}}{\Delta \sigma_i^{\text{exp}}} \right]^2 \quad (8)$$

Lower RMSE, χ^2 , and χ^2/N values indicate closer agreement between the calculated and experimental excitation functions.

The activity of the produced radionuclide was calculated using the medical isotope production formalism implemented in TALYS (Koning et al., 2023),

$$A_i(t) = \lambda_i N_i(t) \quad (8)$$

where $N_i(t)$ is the number of radioactive nuclei produced at time t and λ_i is the corresponding decay constant.

For thick-target irradiation, the incident particle continuously loses energy while traversing the target material. Consequently, the production yield depends not only on the excitation function but also on the stopping power and the effective energy interval within the target. The calculated excitation functions were therefore employed in the TALYS medical isotope production module to calculate the total initial production yield over the investigated projectile-energy range.

After completion of the statistical comparison, the level-density model exhibiting the lowest RMSE, χ^2 , and χ^2/N values was selected for production-yield calculations. The corresponding excitation functions were subsequently employed in the TALYS medical isotope production module to calculate the total initial production yield (GBq/mAh) for both reactions. This procedure provided a consistent framework for comparing the excitation functions, statistical model performance, and practical production capabilities of the two investigated reaction routes.

3. RESULTS

The calculated ground-state, total and cumulative excitation functions for the $^{89}\text{Y}(p,n)^{89}\text{Zr}$ reaction obtained using the CTM, BSFGM, and GSM level-density models are compared with the available experimental data in Figure 1. Overall, all three models reproduce the characteristic shape of the excitation function, including the reaction threshold, the rapid increase in cross section, the maximum production region, and the subsequent decrease at higher proton energies. Nevertheless, noticeable differences are observed among the investigated level-density models, particularly in the vicinity of the peak cross-section region, where the influence of the adopted level-density description on the statistical decay process becomes more pronounced.

The quantitative comparison presented in Table 1 provides a clear assessment of the predictive performances of the investigated level-density models. The statistical indicators (RMSE, χ^2 and χ^2/N) demonstrate that the cumulative excitation functions consistently provide better agreement with the experimental data than the ground-state calculations. Among all investigated calculations, the CTM produced the lowest combined statistical values indicating the best overall agreement

with the available experimental datasets for the $^{89}\text{Y}(p,n)^{89}\text{Zr}$ reaction. The BSFGM and GSM calculations also reproduced the experimental excitation functions reasonably well but exhibited slightly larger statistical deviations.

The calculated excitation functions for the $^{89}\text{Y}(d,2n)^{89}\text{Zr}$ reaction are presented in Figure 2 together with the available experimental datasets. Similar to the proton-induced reaction, the three investigated level-density models reproduce the general behaviour of the experimental excitation functions. However, the deuteron-induced reaction exhibits a broader excitation-function profile because of the more complex reaction mechanisms associated with deuteron interactions, including breakup and pre-equilibrium emission.

The statistical comparison summarized in Table 2 reveals a different ranking of the investigated models for the deuteron-induced reaction. In this case, the BSFGM calculation provides the closest agreement with the experimental data, yielding the lowest combined statistical indicators. The GSM calculation gives comparable results, whereas the CTM calculations produce larger deviations from the experimental measurements.

The statistical evaluation demonstrates that visual agreement between calculated and experimental excitation functions alone is insufficient for selecting the most appropriate nuclear level-density model. Although the excitation functions calculated using the CTM, BSFGM and GSM exhibit similar overall trends, quantitative statistical analysis clearly differentiates their predictive performances. RMSE provides a direct measure of the average deviation between calculated and experimental cross sections, whereas χ^2 and χ^2/N additionally account for the overall distribution of deviations throughout the complete experimental dataset. Therefore, simultaneous evaluation of these statistical indicators provides a substantially

more reliable basis for model selection than qualitative inspection of excitation-function plots alone.

The statistical results further demonstrate that the optimum level-density model depends on the reaction channel rather than a single universal model. For the proton-induced $^{89}\text{Y}(p,n)^{89}\text{Zr}$ reaction, the CTM calculation provides the best overall description of the experimental data, whereas for the deuteron-induced $^{89}\text{Y}(d,2n)^{89}\text{Zr}$ reaction, the BSFGM calculation exhibits the highest predictive accuracy. These results indicate that reaction-specific statistical validation is essential for reliable theoretical modelling of medical radionuclide production.

Based on these statistical results, the CTM was employed in the TALYS medical isotope production module to calculate the total initial production yield (GBq/mAh) for the $^{89}\text{Y}(p,n)^{89}\text{Zr}$ reaction, whereas the BSFGM was adopted for the corresponding calculations of the total initial production yield for the $^{89}\text{Y}(d,2n)^{89}\text{Zr}$ reaction. The calculated total initial production yields are presented in Figure 3. For both production routes, the total initial production yield increases with incident particle energy until reaching the optimum energy region and subsequently decreases as the excitation function declines at higher energies. Consequently, the total initial production yield curves closely follow the behaviour of the corresponding excitation functions and provide practical guidance for selecting the optimum irradiation energy for routine cyclotron production of ^{89}Zr .

Table 1. Statistical comparison (RMSE and χ^2/N) of the investigated level-density models for the $^{89}\text{Y}(p,n)^{89}\text{Zr}$ reaction. Lower statistical values indicate better agreement between the calculated and experimental excitation functions. (N=89 experimental data points)

Model	RMSE (mb)	χ^2	χ^2/N
CTM-Cumulative	64.2312	1395.31	15.6776
BSFGM-Cumulative	76.4395	1440.31	16.1832
GSM-Cumulative	79.5147	1474.16	16.5636
CTM-Total	72.432	1573.79	17.683
GSM-Total	90.3357	1728.80	19.4248
BSFGM-Total	87.42	1788.21	20.0922

Table 2. Statistical comparison (RMSE and χ^2/N) of the investigated level-density models for the $^{89}\text{Y}(d,2n)^{89}\text{Zr}$ reaction. Lower statistical values indicate better agreement between the calculated and experimental excitation functions. (N=47 experimental data points)

Model	RMSE (mb)	χ^2	χ^2/N
BSFGM-Cumulative	40.005	387.142	8.23706
BSFGM-Total	40.891	400.567	8.5227
GSM-Cumulative	42.8694	407.396	8.66799
GSM-Total	48.4384	428.127	9.12067
CTM-Cumulative	54.3972	438.127	9.32186
CTM-Total	53.739	449.357	9.56078

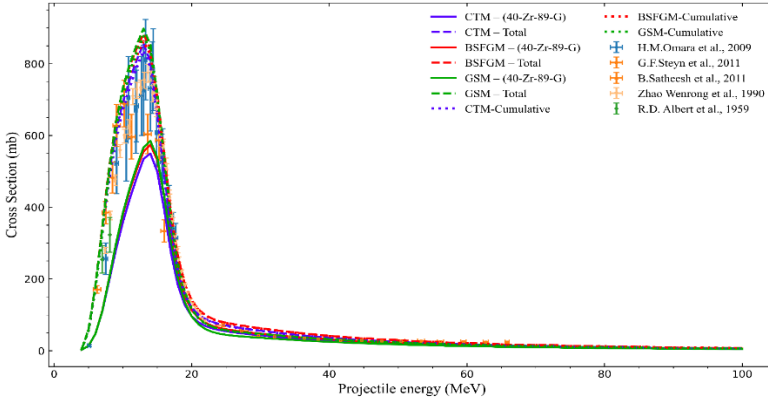


Fig. 1. Calculated ground-state, total, and cumulative excitation functions for the $^{89}\text{Y}(\text{p},\text{n})^{89}\text{Zr}$ reaction obtained using the CTM, BSFGM, and GSM level-density models, compared with the available experimental data.

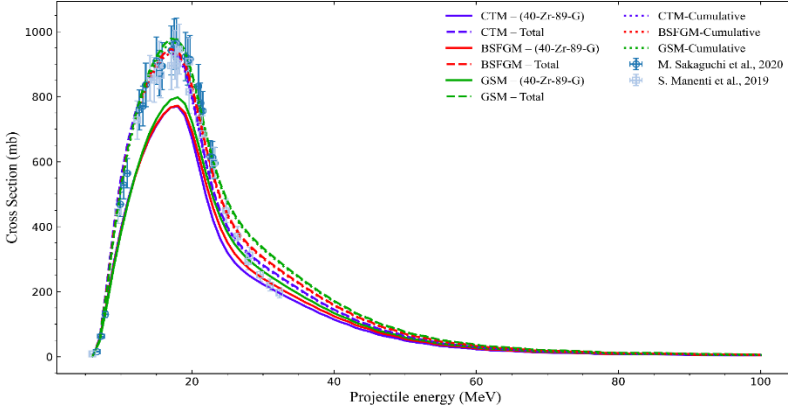


Fig. 2. Calculated ground-state, total, and cumulative excitation functions for the $^{89}\text{Y}(\text{d},2\text{n})^{89}\text{Zr}$ reaction obtained using the CTM, BSFGM, and GSM level-density models, compared with the available experimental data.

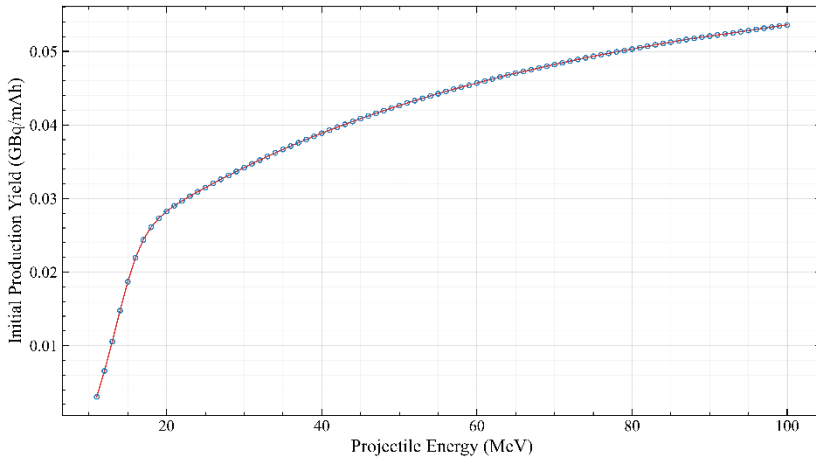


Fig. 3. Calculated total initial production yield of ^{89}Zr as a function of projectile energy for the $^{89}\text{Y}(p,n)^{89}\text{Zr}$ reaction.

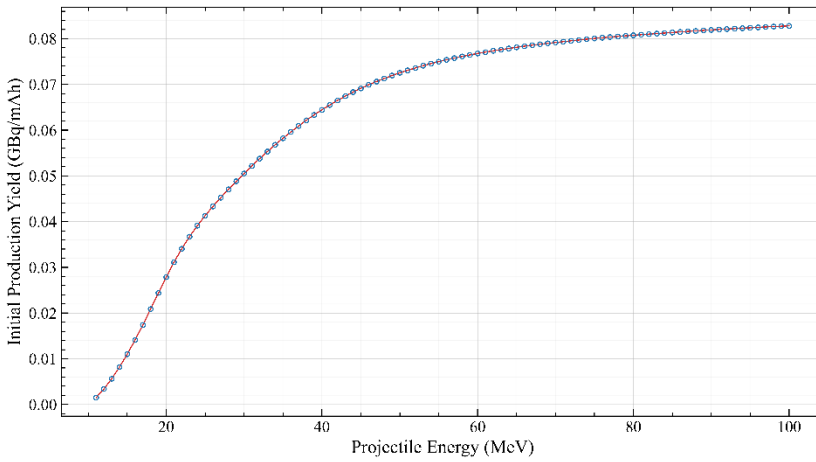


Fig. 4. Calculated total initial production yield of ^{89}Zr as a function of projectile energy for the $^{89}\text{Y}(d,2n)^{89}\text{Zr}$ reaction.

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COMPARATIVE TALYS ANALYSIS OF MEDICAL ^{120}I PRODUCTION VIA THE $^{120}\text{Te}(\text{p},\text{n})$ AND $^{122}\text{Te}(\text{p},3\text{n})$ REACTIONS

Murat DAĞ¹

1. INTRODUCTION

^{120}I is a positron-emitting radioiodine isotope that has attracted considerable interest in nuclear medicine because its physical decay characteristics are well suited for PET. Its half-life allows efficient imaging of rapidly evolving biological processes, while the chemical properties of iodine facilitate its incorporation into biologically active molecules and radiopharmaceutical compounds. Consequently, ground-state ^{120}I has emerged as a promising radionuclide for short-duration PET imaging, pharmacokinetic investigations, and the development of radioiodine-labelled tracers (Herzog et al., 2006; Qaim et al., 2011).

The medical importance of ground-state ^{120}I is closely associated with its favourable decay characteristics and its applicability to quantitative PET imaging. Early investigations established the fundamental nuclear properties of iodine and xenon isotopes around mass number 120 (Butement & Qaim, 1965), whereas subsequent studies provided accurate decay data and positron-emission intensities required for reliable activity determination and quantitative PET applications (Hohn et al., 2000). These studies have established the nuclear data required

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for both theoretical production calculations and practical medical applications of ground-state ^{120}I .

Cyclotron production of ground-state ^{120}I is commonly performed using enriched tellurium targets. Among the available proton-induced production routes, the $^{120}\text{Te}(p,n)^{120}\text{I}$ reaction is the most widely investigated because of its relatively low threshold energy and compatibility with conventional medical cyclotrons (Hohn et al., 1998a). In contrast, the $^{122}\text{Te}(p,3n)^{120}\text{I}$ reaction represents an alternative production route that requires substantially higher proton energies owing to the emission of three neutrons but is capable of providing higher production yields under suitable irradiation conditions (Hohn et al., 1998b). Experimental investigations have demonstrated that both reactions are feasible production routes, although they differ considerably in terms of optimum proton-energy window, production yield, and radionuclidic purity (Qaim et al., 2003; El-Azony et al., 2008).

Reliable prediction of excitation functions is essential for optimizing irradiation conditions, estimating production performance, and maximizing the practical production of medical radionuclides before experimental irradiation. Modern nuclear reaction codes provide an efficient theoretical framework for performing such investigations. Among these codes, TALYS has become one of the most widely used computational tools for calculating excitation functions, reaction cross sections, radionuclide activities, and production yields over a broad range of target nuclei and incident particles (Koning et al., 2023; Koning & Rochman, 2012). Nevertheless, the predictive capability of TALYS calculations depends strongly on the adopted nuclear models, particularly the nuclear level-density model governing the statistical decay of the compound nucleus.

Nuclear level density is one of the most influential quantities in statistical nuclear reaction calculations because it governs particle-emission probabilities and therefore directly affects the calculated excitation functions. Among the phenomenological formulations, the Constant Temperature Model (CTM), the Back-Shifted Fermi Gas Model (BSFGM), and the Generalized Superfluid Model (GSM) are the approaches most frequently employed for charged-particle-induced reaction calculations (Gilbert & Cameron, 1965; Ignatyuk et al., 1979). Although these models have been successfully applied to numerous medical radionuclide production studies, their predictive performance may vary depending on the target nucleus and reaction channel. Consequently, systematic comparison of these models using available experimental excitation-function data remains important for improving the reliability of theoretical production calculations.

Although numerous experimental and theoretical investigations have examined both proton-induced production routes leading to ground-state ^{120}I , most studies have primarily focused on reproducing excitation functions, determining maximum cross sections, or reporting activity and production yields (Sudar et al., 2000; Qaim et al., 2003). Comparatively fewer studies have systematically assessed the predictive performance of different level-density models using quantitative statistical criteria while simultaneously comparing both production routes within a common theoretical framework. Such an evaluation is particularly valuable because visual agreement alone may not always identify the model that most accurately reproduces the available experimental data.

In the present study, the principal proton-induced production pathways for ground-state ^{120}I were investigated using the TALYS nuclear reaction code with three phenomenological nuclear level-density models: CTM, BSFGM, and GSM. Ground-

state excitation functions were calculated and compared with the available experimental datasets. The predictive performance of each level-density model was quantitatively evaluated using the root mean square error (RMSE), chi-square (χ^2), and chi-square per experimental point (χ^2/N). Finally, the statistically best-performing level-density model for each reaction was employed in the TALYS medical isotope production module to calculate the activity and the initial production yield of ground-state ^{120}I as a function of the incident proton energy. This combined evaluation provides a consistent theoretical framework for comparing the two production routes and for identifying the most suitable modelling approach for theoretical production of ground-state ^{120}I .

2. METHODS

The theoretical production of ground-state ^{120}I via two proton-induced tellurium reaction pathways was investigated using the TALYS nuclear reaction code. These reactions were selected because they represent the principal low- and high-energy proton-induced production routes for ground-state ^{120}I and provide an appropriate basis for comparing different reaction mechanisms under identical theoretical conditions (Hohn et al., 1998a; Hohn et al., 1998b). The $^{120}\text{Te}(p,n)$ reaction proceeds predominantly through single-neutron emission, whereas the $^{122}\text{Te}(p,3n)$ reaction involves three-neutron emission and therefore requires substantially higher incident proton energies.

The TALYS nuclear reaction code was employed as the computational tool for modelling proton-induced nuclear reactions in tellurium targets. The code combines optical-model calculations, direct reactions, pre-equilibrium emission, compound-nucleus formation, and statistical decay within a unified theoretical framework, allowing the calculation of

excitation functions, residual radionuclide production cross sections, radionuclide activities, and production yields over a broad range of target nuclei with mass numbers greater than or equal to 12 (Koning et al., 2023; Koning & Rochman, 2012).

All calculations were performed under identical computational conditions for the two investigated reactions. The incident proton energy was varied over the energy region relevant to medical cyclotron production, whereas the nuclear level-density model was treated as the principal variable in order to evaluate its influence on the calculated excitation functions.

For each production route, ground-state excitation functions were calculated as a function of incident proton energy. These calculations were used to determine the reaction-threshold behaviour, the energy dependence of ground-state ^{120}I formation, the maximum production cross section, and the proton energy corresponding to this maximum. Comparison of the two excitation functions provided a common theoretical basis for evaluating the low-energy single-neutron and high-energy three-neutron production routes.

A central component of the theoretical calculations is the treatment of nuclear level density. In TALYS, the level density $\rho(E_x, J, \Pi)$ represents the number of nuclear levels per unit energy interval at excitation energy E_x for spin J and parity Π . The total level density is obtained by summing the contributions from all spin and parity states,

$$\rho_{\text{tot}}(E_x) = \sum_{J, \Pi} \rho(E_x, J, \Pi). \quad (1)$$

A commonly adopted analytical description of the level density is provided by the Fermi-gas model (Gilbert & Cameron, 1965),

$$\rho_F^{\text{tot}}(E_x) = \frac{\sqrt{\pi} \exp(2\sqrt{a}U)}{12\sqrt{2} a^{1/4} U^{5/4} \sigma} = \frac{\omega_F^{\text{tot}}(E_x)}{\sqrt{2\pi}\sigma} \quad (2)$$

where U denotes the effective excitation energy, σ is the spin cut-off parameter, and a is the level-density parameter. In early formulations, the level-density parameter was generally treated as energy independent. To account for shell effects, Ignatyuk and co-workers introduced an excitation-energy-dependent level-density parameter expressed as

$$a(E_x) = \tilde{a} \left[1 + \delta W \frac{1 - \exp(-\gamma U)}{U} \right] \quad (3)$$

where the asymptotic level-density parameter is

$$\tilde{a} = \alpha A + \beta A^{2/3}. \quad (4)$$

In these expressions, δW is the shell-correction energy, γ is the shell-damping parameter, and α and β are global constants adjusted to reproduce average level-density systematics (Ignatyuk et al., 1979).

Three phenomenological level-density models implemented in TALYS were investigated in this study: CTM, BSFGM, and GSM. CTM assumes a constant nuclear temperature at low excitation energies and connects this region to a Fermi-gas description above a matching energy. The BSFGM applies the Fermi-gas formalism while accounting for pairing effects through an empirical back-shift parameter. The GSM incorporates pairing correlations based on the superfluid model and describes the transition from the superfluid regime at low excitation energies to the normal Fermi-gas region at higher excitation energies.

These models were selected because they are among the most widely used phenomenological level-density approaches available in TALYS and have been extensively applied in charged-particle-induced reaction calculations. Their comparison makes it possible to evaluate the sensitivity of the calculated ground-state ^{120}I excitation functions to the adopted statistical description of compound-nucleus decay.

Experimental ground-state excitation-function data for both investigated production routes were compiled from the EXFOR database together with the corresponding original publications (Otuka et al., 2014). Only experimental measurements explicitly associated with the ground-state production of ^{120}I were included in the statistical comparison. The available datasets were used to evaluate the predictive capability of CTM, BSFGM, and GSM calculations over the investigated proton-energy ranges.

For each experimental data point, the theoretical cross section was obtained at the corresponding proton energy by interpolation of the calculated excitation function. The predictive performance of each level-density model was evaluated using the root mean square error (RMSE), chi-square (χ^2), and chi-square per experimental point (χ^2/N). The RMSE was calculated as

$$\text{RMSE} = \sqrt{\frac{1}{N} \sum_{i=1}^N (\sigma_i^{\text{calc}} - \sigma_i^{\text{exp}})^2} \quad (5)$$

where N is the total number of experimental data points, and σ^{calc} and σ^{exp} are the calculated and experimental cross sections, respectively. The chi-square value was calculated as

$$\chi^2 = \sum_{i=1}^N \left[\frac{\sigma_i^{\text{calc}} - \sigma_i^{\text{exp}}}{\Delta\sigma_i^{\text{exp}}} \right]^2 \quad (6)$$

where $\Delta\sigma_i^{\text{exp}}$ denotes the experimental cross-section uncertainty. The chi-square value per experimental point was then obtained from

$$\frac{\chi^2}{N} = \frac{1}{N} \sum_{i=1}^N \left[\frac{\sigma_i^{\text{calc}} - \sigma_i^{\text{exp}}}{\Delta\sigma_i^{\text{exp}}} \right]^2 \quad (7)$$

Lower RMSE, χ^2 , and χ^2/N values indicate closer agreement between the calculated and experimental excitation functions.

The activity of ground-state ^{120}I was calculated using the medical isotope production formalism implemented in TALYS (Koning et al., 2023). In this formalism, the number of produced radioactive nuclei is followed during irradiation by considering both continuous radionuclide production and radioactive decay. The activity of radionuclide i at time t is expressed as

$$A_i(t) = \lambda_i N_i(t) \quad (8)$$

where $N_i(t)$ is the number of radioactive nuclei present at time t and λ_i is the corresponding decay constant. The calculated activity–time curves were used to describe the build-up of ground-state ^{120}I during irradiation and its gradual approach to saturation.

For thick-target irradiation, the incident proton continuously loses energy while traversing the enriched tellurium target. Consequently, the production yield depends not only on the magnitude of the excitation function but also on the stopping power of the target material and the effective proton-energy

interval within the target. The calculated ground-state excitation functions were therefore employed in the TALYS medical isotope production module to determine the initial production yield of ground-state ^{120}I as a function of incident proton energy.

The initial production yield was expressed in GBq/mAh and was evaluated over the investigated proton-energy range for each reaction. This quantity represents the activity produced per unit integrated beam current under the adopted TALYS production conditions and provides a practical measure for comparing the production capabilities of the low-energy $^{120}\text{Te}(p,n)$ and high-energy $^{122}\text{Te}(p,3n)$ routes.

After completion of the statistical comparison, the level-density model exhibiting the lowest RMSE, χ^2 , and χ^2/N values for each reaction was selected for the subsequent activity and initial-production-yield calculations. The corresponding ground-state excitation functions were used to calculate the activity as a function of irradiation time and the initial production yield as a function of incident proton energy. This procedure provided a consistent framework for comparing the excitation functions, statistical model performance, activity behaviour, and practical production capabilities of the two investigated ground-state ^{120}I production routes.

3. RESULTS

The calculated excitation functions for the $^{120}\text{Te}(p,n)^{120}\text{I}$ reaction obtained using CTM, BSFGM, and GSM level-density models are compared with the available experimental data in Figure 1. Overall, all three models reproduce the characteristic shape of the excitation function, including the reaction threshold, the rapid increase in cross section, the maximum production region, and the subsequent decrease at higher proton energies. Nevertheless, noticeable differences are observed among the

investigated level-density models, particularly in the vicinity of the peak cross-section region, where the influence of the adopted level-density description on the statistical decay process becomes more pronounced.

The quantitative comparison presented in Table 1 provides a clear assessment of the predictive performances of the investigated level-density models. The statistical indicators (RMSE, χ^2 and χ^2/N) demonstrate that CTM calculation provides the best overall agreement with the available experimental datasets for the $^{120}\text{Te}(p,n)^{120}\text{I}$ reaction. The BSFGM and GSM calculations also reproduce the experimental excitation function reasonably well but exhibit larger statistical deviations from the experimental measurements. Overall, the statistical analysis indicates that CTM offers the most reliable theoretical description of the available experimental data for this reaction.

The calculated excitation functions for the $^{122}\text{Te}(p,3n)^{120}\text{I}$ reaction are presented in Figure 2 together with the available experimental datasets. Similar to the $^{120}\text{Te}(p,n)^{120}\text{I}$ reaction, the three investigated level-density models reproduce the general behaviour of the experimental excitation function over the investigated proton-energy range. However, the $^{122}\text{Te}(p,3n)^{120}\text{I}$ reaction exhibits a broader excitation-function profile because of its higher reaction threshold and the emission of three neutrons. Small differences among the investigated models become more apparent around the maximum cross-section region, reflecting the sensitivity of the statistical decay calculations to the adopted level-density description.

The statistical comparison summarized in Table 2 reveals that CTM calculation again provides the closest agreement with the available experimental data. Among the investigated level-density models, CTM yields the lowest RMSE, χ^2 and χ^2/N values, indicating the highest predictive performance for the

$^{122}\text{Te}(p,3n)^{120}\text{I}$ reaction. The BSFGM and GSM calculations also reproduce the overall behaviour of the excitation function but exhibit progressively larger statistical deviations throughout the experimental energy range.

The statistical evaluation demonstrates that visual agreement between calculated and experimental excitation functions alone is insufficient for selecting the most appropriate nuclear level-density model. Although the excitation functions calculated using CTM, BSFGM and GSM exhibit similar overall trends for both investigated reactions, quantitative statistical analysis clearly differentiates their predictive performances. RMSE provides a direct measure of the average deviation between calculated and experimental cross sections, whereas χ^2 and χ^2/N additionally account for the overall distribution of deviations throughout the complete experimental dataset. Therefore, simultaneous evaluation of these statistical indicators provides a substantially more reliable basis for model selection than qualitative inspection of excitation-function plots alone.

Although CTM provides the best overall statistical performance for both investigated production routes, the relative differences among the three level-density models are not identical for the two reactions. For the $^{120}\text{Te}(p,n)$ route, all investigated models produce comparatively similar RMSE values, indicating that each model reproduces the measured excitation function with a similar average deviation. In contrast, the χ^2 and χ^2/N values exhibit a clearer separation, demonstrating that CTM achieves a more consistent description over the entire experimental energy range rather than only around the maximum cross section. A different behaviour is observed for the $^{122}\text{Te}(p,3n)$ route, where the statistical differences between the models become considerably larger. This increased separation indicates that the predictive capability of the calculations is more sensitive to the adopted level-density description for the higher-energy reaction

channel, making model selection substantially more influential for reliable theoretical predictions.

The statistical results further demonstrate that CTM consistently provides the best overall agreement with the available experimental data for both investigated production routes. These findings indicate that the Constant Temperature Model provides the most suitable level-density description among the investigated models for theoretical calculations of both proton-induced production routes leading to ground-state ^{120}I .

Based on these statistical results, CTM was employed in the TALYS medical isotope production module to calculate the activity and the initial production yield (GBq/mAh) of ground-state ^{120}I for both investigated reaction channels. The calculated initial production yields of ground-state ^{120}I are presented in Figures 3 and 4. For both production routes, the initial production yield increases with incident proton energy until reaching the optimum energy region and subsequently decreases as the excitation function declines at higher energies. Consequently, the calculated initial production yield curves closely follow the behaviour of the corresponding excitation functions and provide practical guidance for selecting the optimum irradiation energy for the routine cyclotron production of ground-state ^{120}I .

The marked difference between the statistical values obtained for the two target isotopes shows that the quality of model reproduction is strongly dependent on the reaction channel. For the lower-energy route, the three calculations remain relatively close to one another and to the experimental measurements, as reflected by the comparatively small RMSE values. In contrast, the high-energy three-neutron channel displays substantially larger discrepancies, particularly in the peak region, where the measured cross sections exceed the calculated curves. This behaviour suggests that the description of

multi-neutron emission is more demanding than that of the single-neutron channel and that level-density selection alone cannot account for the entire difference between calculation and experiment.

The yield curves further distinguish the practical behaviour of the two routes. Production from the ^{120}Te target rises rapidly within the lower proton-energy region and then increases more gradually, whereas irradiation of ^{122}Te requires a higher incident energy before a comparable rise in yield is observed. The latter route ultimately reaches a larger calculated production level, but this advantage is accompanied by the need for a higher-energy proton beam. Accordingly, the choice between the two pathways should not be based solely on the maximum calculated yield; accelerator capability, target enrichment, usable energy range, and the required production scale should also be considered when planning ground-state ^{120}I production.

Table 1. Statistical comparison (RMSE and χ^2/N) of the investigated level-density models for the $^{120}\text{Te}(\text{p},\text{n})^{120}\text{I}$ reaction. Lower statistical values indicate better agreement between the calculated and experimental excitation functions. (N=40 experimental data points)

Model	RMSE (mb)	χ^2	χ^2/N
CTM	11.0676	89.217	2.23043
BSFGM	10.7995	109.929	2.74823
GSM	11.7242	142.685	3.56712

Table 2. Statistical comparison (RMSE and χ^2/N) of the investigated level-density models for the $^{122}\text{Te}(p,3n)^{120}\text{I}$ reaction. Lower statistical values indicate better agreement between the calculated and experimental excitation functions. (N=13 experimental data points)

Model	RMSE (mb)	χ^2	χ^2/N
CTM	186.671	152.283	11.7141
BSFGM	196.778	174.405	13.4158
GSM	206.635	189.61	14.5853

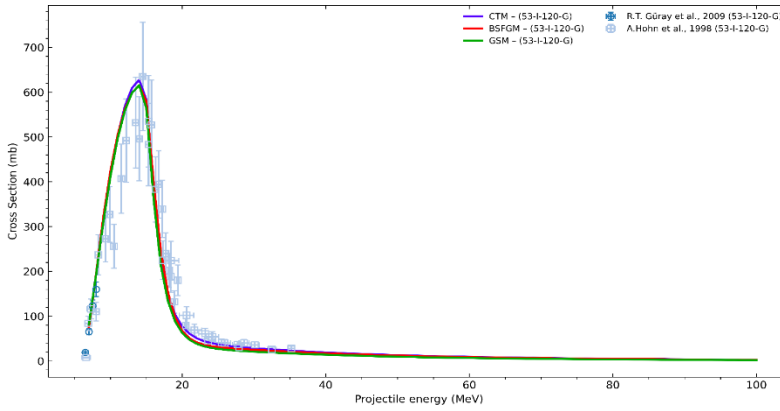


Fig. 1. Calculated ground-state excitation functions for the $^{120}\text{Te}(p,n)^{120}\text{I}(\text{ground})$ reaction obtained using CTM, BSFGM, and GSM level-density models, compared with the available experimental data.

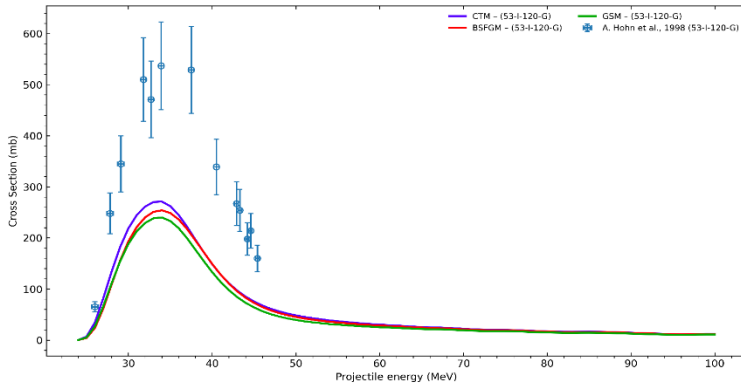


Fig. 2. Calculated ground-state excitation functions for the $^{122}\text{Te}(p,3n)^{120}\text{I}$ reaction obtained using CTM, BSFGM, and GSM level-density models, compared with the available experimental data.

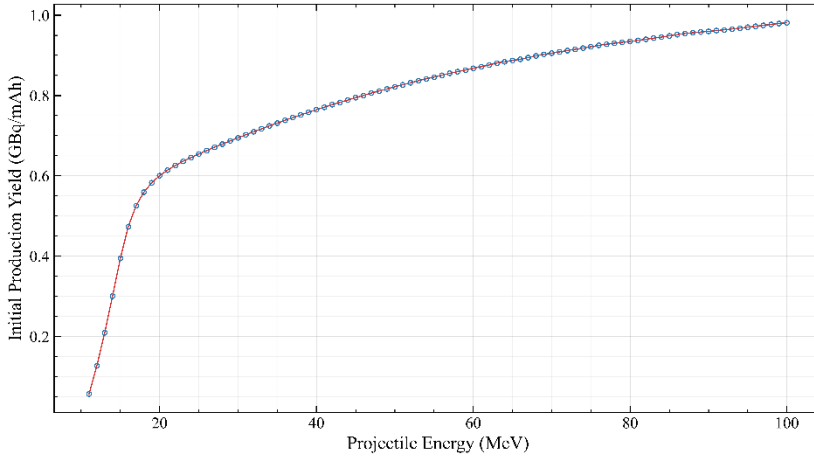


Fig. 3. Calculated initial production yield of ground state of ^{120}I as a function of projectile energy for the $^{120}\text{Te}(p,n)^{120}\text{I}$ reaction.

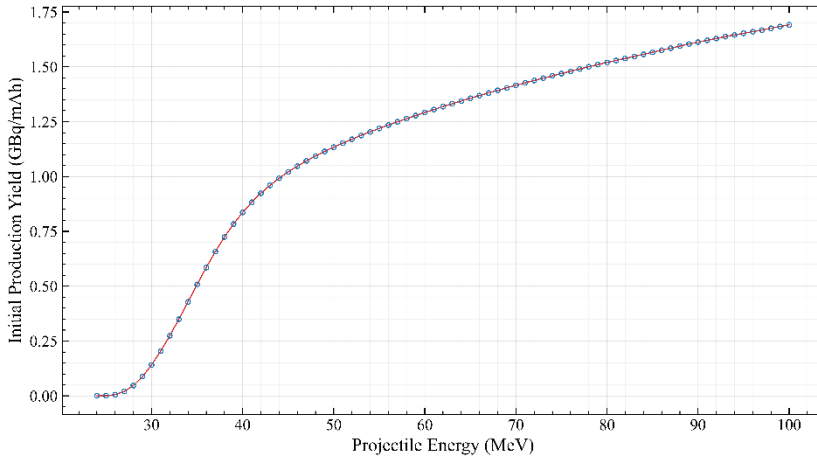


Fig. 4. Calculated initial production yield of ground state of ^{120}I as a function of projectile energy for the $^{122}\text{Te}(p,3n)^{120}\text{I}$ reaction.

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BaO KATKILI PLA KOMPOZİTLERİNİN RADYASYON ZIRHLAMA ÖZELLİKLERİNİN XCOM VE Phy-X/PSD İLE İNCELENMESİ

Seda ERDÖNMEZ¹

1. GİRİŞ

Radyasyon, elektromanyetik dalga veya parçacık biçiminde enerjinin yayılması ya da aktarılmasıdır. Radyasyon, maddeyi iyonlaştırma yeteneğine göre iyonlaştırıcı ve iyonlaştırıcı olmayan radyasyon olmak üzere iki temel kategoriye ayrılabilir. İyonlaştırıcı olmayan radyasyon, etkileştiği atom veya moleküllerden elektron kopararak iyonlaşmaya neden olacak kadar yüksek enerjiye sahip değildir. Radyo dalgaları, mikrodalgalar, kızılötesi ışınlar, görünür ışık ve morötesi ışığın büyük bölümü iyonlaştırıcı olmayan radyasyon grubunda yer almaktadır. Madde ile etkileşimi sonucunda doğrudan veya dolaylı olarak iyonlaşmaya neden olabilecek yeterli enerjiye sahip radyasyon türleri ise iyonlaştırıcı radyasyon olarak adlandırılmaktadır. X-ışınları ve gama ışınları ile alfa ve beta parçacıkları ve nötronlar iyonlaştırıcı radyasyon türleri arasında yer almaktadır (Little, 2003; More, Alsayed, Badawi, Thabet, & Pawar, 2021; Uosif, 2014).

İyonlaştırıcı radyasyonun insan sağlığı üzerindeki etkileri uzun yıllardır bilinmektedir. Wilhelm Röntgen tarafından 1895 yılında X-ışınlarının keşfedilmesinden kısa bir süre sonra, X-ışınlarına maruz kalan bireylerde cilt yanıkları ve radyasyona bağlı diğer deri hasarları rapor edilmiştir. Takip eden yıllarda ise

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radyasyon maruziyetine bağlı cilt kanseri vakaları bildirilmiştir (Little, 2003) . Günümüzde iyonlaştırıcı radyasyonun DNA hasarı, kalıtsal etkiler, doku ve organ hasarı ile kanser riskinde artış gibi çeşitli sağlık sorunlarına yol açabileceği bilinmektedir (Christensen, Iddins, & Sugarman, 2014; Little, 2003; Ron, 1998). İyonlaştırıcı radyasyonun zararlı etkilerinden korunmada kullanılan temel yöntemlerden biri uygun radyasyon zırhlayıcı malzemelerin kullanılmasıdır. Kurşun ve beton, yüksek radyasyon zayıflatma özellikleri nedeniyle uzun yıllardır zırhlama amacıyla yaygın olarak kullanılmaktadır. Bununla birlikte, kurşunun yüksek yoğunluğu ve toksik yapısı, betonun ise yüksek ağırlığı ve hacimli yapısı kullanım açısından çeşitli dezavantajlar oluşturmaktadır (Al-Buriahi et al., 2021). Bu nedenle araştırmacılar, geleneksel zırhlayıcı malzemelere alternatif olabilecek daha hafif, toksik olmayan ve kolay işlenebilir malzemelerin geliştirilmesine yönelik çalışmalar yürütmektedir.

Polimerler; düşük yoğunlukları, düşük üretim maliyetleri, kolay işlenebilirlikleri ve farklı geometrilere şekillendirilebilmeleri gibi avantajları nedeniyle kurşun esaslı geleneksel malzemelere alternatif olabilecek radyasyon zırhlayıcı malzeme adayları arasında yer almaktadır. Bununla birlikte, polimerlerin yapısında çoğunlukla düşük atom numaralı elementlerin bulunması nedeniyle katkısız halde radyasyon zayıflatma performansları genellikle sınırlıdır. Bu nedenle polimerlerin radyasyon zırhlama performanslarının geliştirilmesi amacıyla yüksek atom numarasına sahip elementler veya bu elementlerin bileşikleri polimer matrislere katkı maddesi olarak ilave edilmektedir (Ambika, Nagaiah, & Prashantha, 2020).

Bilimsel literatür incelendiğinde, farklı polimer matrislerin radyasyon zırhlama özelliklerinin geliştirilmesi amacıyla çeşitli katkı maddeleri ile modifiye edildiği görülmektedir. Epoksi (R. Li et al., 2017) , polivinil alkol (PVA)

(Hosseini, Malekie, & Kazemi, 2022) , düşük yoğunluklu polietilen (LDPE) (Alshahri et al., 2021) , yüksek yoğunluklu polietilen (HDPE) (El-Sayed Abdo, Ali, & Ismail, 2003) , poliüretan (PU) (Knott et al., 2023) , polikarbonat (PC) (Mirji & Lobo, 2020) , polietilen tereftalat (PET) (Aygün & Alma, 2020) , polistiren (PS) (Jaiyen, Phunpueok, & Thongpool, 2019) , polimetil metakrilat (PMMA) (Z. Li et al., 2016) , polivinil klorür (PVC) (Gholamzadeh, Sharghi, & Aminian, 2022) , polivinilpirolidon (PVP) (Abdali, 2022) ve polipropilen (PP) (Ekinci et al., 2022) bu amaçla araştırılan polimerlerden bazılarıdır. Bunun yanı sıra tungsten (Hosseini et al., 2022) , bizmut (Alshahri et al., 2021) , baryum (Jaiyen et al., 2019) , antimon (Sayyed, Tekin, Altunsoy, Obaid, & Almatari, 2018) , seryum (Yalcin, Aktas, & Yilmaz, 2019) ve gadolinyum (R. Li et al., 2017) gibi yüksek atom numaralı elementler ve bunların bileşikler, radyasyon zayıflatma performansını artırmak amacıyla polimer matrislerde katkı maddesi olarak kullanılmaktadır.

Örneğin Kılıç ve ark., PbO ve BaO ile katkılандırılmış epoksi reçinelerin gama ışını zırhlama performanslarını NaI(Tl) dedektörü ve dar ışın geometrisine sahip deneysel bir ölçüm sistemi kullanarak araştırmışlardır (Kılıç, Ergin, Karabul, & Güven Özdemir, 2019). Çalışmada kütle azaltma katsayısı, yarı değer kalınlığı, onda bir değer kalınlığı ve ortalama serbest yol gibi radyasyon zırhlama parametreleri değerlendirilmiştir. Aynı katkı oranlarında PbO ve BaO içeren epoksi kompozitler karşılaştırıldığında, PbO içeren kompozitlerin kütle azaltma katsayılarının BaO içeren kompozitlere göre daha yüksek olduğu belirlenmiştir. Bununla birlikte, BaO içeren kompozitler arasında en yüksek BaO oranına sahip Epoksi/%40 BaO kompozitinin 81 ve 356 keV foton enerjilerindeki kütle azaltma katsayılarının çelik, tuğla, çimento, beton ve Epoksi/Gd₂O₃ kompozitlerinden daha yüksek olduğu bildirilmiştir. Başka bir çalışmada Beyazay

ve ark., PCz/BaO nanokompozitlerinin iyonlaştırıcı radyasyon zırhlama performanslarını deneysel yöntemler ve WinXCom kullanarak incelemiş ve en yüksek BaO katkı oranına sahip kompozitin özellikle 81 keV foton enerjisinde en yüksek zırhlama performansını gösterdiğini belirlemişlerdir (Beyazay, Karabul, Korkut, Kılıç, & Özdemir, 2022).

Bu çalışmada PLA esaslı BaO katkılı kompozitlerin radyasyon zırhlama performanslarının teorik olarak incelenmesi amaçlanmıştır. Polilaktik asit (PLA) kompozitlerin matris bileşeni olarak, BaO ise yapısında yüksek atom numarasına sahip baryum ($Z = 56$) bulundurması nedeniyle katkı maddesi olarak seçilmiştir. Bu amaçla ağırlıkça %5, %10 ve %20 BaO içerecek şekilde PLA esaslı kompozit bileşimleri tasarlanmıştır. Bu yapıların radyasyon zırhlama parametreleri XCOM ve Phy-X/PSD hesaplama araçları kullanılarak teorik olarak belirlenmiştir. Elde edilen sonuçlar üzerinden BaO katkı oranının kompozitlerin radyasyon zırhlama parametreleri üzerindeki etkisi değerlendirilmiştir.

2. MATERYAL VE METOT

PLA/BaO kompozitlerinin radyasyon zırhlama özelliklerinin değerlendirilmesi amacıyla kütle azaltma katsayısı (Mass attenuation coefficient, MAC), lineer azaltma katsayısı (Linear attenuation coefficient, LAC), yarı değer kalınlığı (Half value layer, HVL) ve etkin atom numarası (Z_{etk}) incelenmiştir. Bu kapsamda kompozitlerin kütle azaltma katsayıları XCOM ile 1 keV–100 GeV enerji aralığında hesaplanmış ve elde edilen MAC değerleri kullanılarak LAC, HVL ve Z_{etk} değerleri belirlenmiştir. Ayrıca, aynı parametreler Phy-X/PSD hesaplama aracı ile de hesaplanmıştır. Hesaplamalarda kullanılan saf PLA ve farklı oranlarda BaO içeren PLA/BaO kompozitlerinin elementel

ağırlık fraksiyonları ile teorik olarak hesaplanan yoğunlukları Tablo 1’de verilmiştir.

Tablo 1. Saf PLA ve PLA/BaO kompozitlerinin elementel ağırlık fraksiyonları ve yoğunlukları

Örnek	H	C	O	Ba	Yoğunluk (g/cm ³)
PLA	0,05595	0,49999	0,44405	-	1,20
PLA-5Ba	0,05316	0,47499	0,42707	0,04478	1,28
PLA-10Ba	0,05036	0,45000	0,41008	0,08957	1,35
PLA-20Ba	0,04476	0,40000	0,37611	0,17913	1,54

2.1. Radyasyon Zırhlama Parametreleri

Beer-Lambert yasasına göre, dar ve monoenerjetik bir foton demeti zırhlayıcı bir malzemeden geçtiğinde ışınım şiddeti, malzeme kalınlığına bağlı olarak üstel biçimde azalmaktadır. Bu ilişki Denklem 1 ile ifade edilmektedir.

$$I = I_0 e^{-\mu x} \quad (1)$$

Burada I_0 malzemeye gelen başlangıç ışınım şiddetini, I malzemeden geçen ışınım şiddetini, x malzeme kalınlığını ve μ lineer azaltma katsayısını ifade etmektedir. Lineer azaltma katsayısı, bir fotonun birim yol uzunluğu başına malzeme ile etkileşme olasılığını ifade eden ve malzemenin radyasyon zayıflatma performansının değerlendirilmesinde kullanılan temel parametrelerden biridir. Lineer azaltma katsayısının malzeme yoğunluğuna bölünmesiyle kütle azaltma katsayısı (MAC) elde edilir (Denklem 2) (Gurler & Akar Tarım, 2012).

$$MAC = \frac{\mu}{\rho} (cm^2 g^{-1}) \quad (2)$$

Burada MAC kütle azaltma katsayısını, μ lineer azaltma katsayısını ve ρ zırhlayıcı malzemenin yoğunluğunu ifade etmektedir.

Yarı değer kalınlığı (HVL), gelen foton ışınımının şiddetini başlangıç değerinin yarısına düşürmek için gerekli

zırhlayıcı malzeme kalınlığı olarak tanımlanmaktadır. HVL değeri lineer azaltma katsayısına bağlı olarak Denklem 3 ile hesaplanmaktadır (Akkaş, 2016).

$$HVL = \frac{\ln 2}{\mu} \quad (3)$$

Düşük HVL değerleri, foton şiddetinin daha küçük bir malzeme kalınlığında yarıya indirilebildiğini ve dolayısıyla malzemenin daha yüksek radyasyon zayıflatma performansına sahip olduğunu göstermektedir.

Farklı elementlerden oluşan bileşik ve kompozit malzemelerin radyasyon zırhlama özelliklerinin değerlendirilmesinde etkin atom numarası (Z_{etk}) kullanılmaktadır. Etkin atom numarası, çok elementli bir malzemenin radyasyonla etkileşim davranışını tek bir eşdeğer atom numarası ile ifade eden bir parametredir. Z_{etk} değeri malzemenin elementel bileşimine ve foton enerjisine bağlı olarak değişebilmektedir ve interpolasyon yöntemi kullanılarak Denklem 4 ile hesaplanabilmektedir (Levet & Özdemir, 2017; Singh, Badiger, & Kucuk, 2014).

$$Z_{etk} = \frac{Z_1(\log \sigma_2 - \log \sigma_{etk}) + Z_2(\log \sigma_{etk} - \log \sigma_1)}{\log \sigma_2 - \log \sigma_1} \quad (4)$$

Burada σ_{etk} , incelenen malzemenin toplam atomik tesir kesitini; σ_1 ve σ_2 , σ_{etk} değerini çevreleyen elementlere ait atomik tesir kesitlerini; Z_1 ve Z_2 ise bu elementlerin atom numaralarını ifade etmektedir. Malzemenin toplam atomik tesir kesiti (σ_{etk}), kütle azaltma katsayısından Denklem 5 kullanılarak hesaplanabilir.

$$\sigma_{etk} = \frac{(\mu/\rho)_{malzeme}}{N_A \sum w_i/A_i} \quad (5)$$

Burada w_i , malzemeyi oluşturan i. elementin ağırlık fraksiyonunu, A_i i. elementin atomik kütesini ve N_A Avogadro sabitini ifade etmektedir (Çağlar et al., 2019).

2.2. XCOM ve Phy-X/PSD Hesaplamaları

Bu çalışmada PLA ve PLA/BaO kompozitlerinin radyasyon zırhlama parametrelerinin teorik olarak belirlenmesinde NIST XCOM veri tabanı ve Phy-X/PSD çevrimiçi hesaplama yazılımı kullanılmıştır.

XCOM (Photon Cross Sections Database), Berger ve Hubbell tarafından geliştirilen ve National Institute of Standards and Technology (NIST) tarafından sunulan bir foton etkileşim veri tabanıdır (Berger & Hubbell, 1987; Gerward, Guilbert, Jensen, & Leving, 2004). XCOM kullanılarak element, bileşik ve karışımlar için 1 keV–100 GeV foton enerji aralığında koherent ve inkoherent saçılma, fotoelektrik soğurma ve çift oluşumu gibi temel foton etkileşimlerine ait tesir kesitleri ile toplam kütle azaltma katsayıları hesaplanabilmektedir. Bileşik ve karışımların kütle azaltma katsayıları, karışım kuralına göre Denklem 6 kullanılarak belirlenmektedir.

$$(MAC)_{malzeme} = \sum_i^n w_i (MAC)_i \quad (6)$$

Burada (w_i), malzemeyi oluşturan i. elementin ağırlık fraksiyonunu, $(MAC)_i$ ise i. elementin ilgili foton enerjisindeki kütle azaltma katsayısını ifade etmektedir.

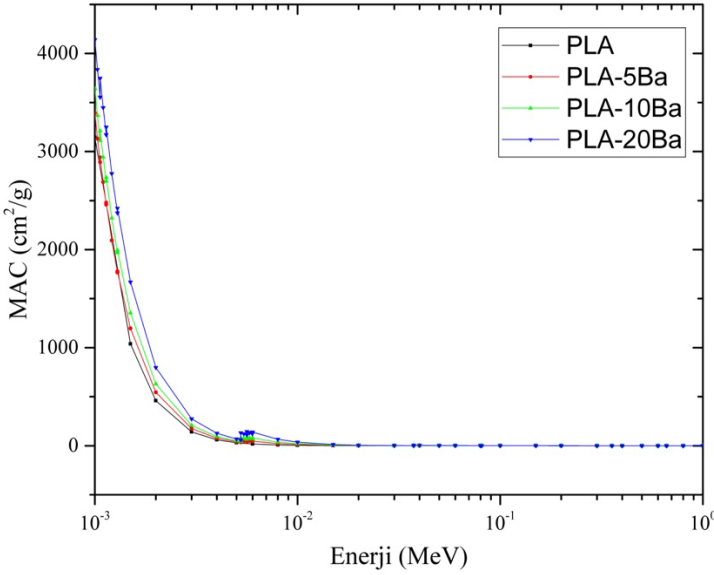
Phy-X/PSD (Photon Shielding and Dosimetry), Şakar ve ark. tarafından radyasyon zırhlama ve dozimetri parametrelerinin hesaplanması amacıyla geliştirilen çevrimiçi bir hesaplama yazılımıdır (Şakar, Özpolat, Alım, Sayyed, & Kurudirek, 2020). Phy-X/PSD ile kütle ve lineer azaltma katsayılarının yanı sıra yarı değer kalınlığı (HVL), onda bir değer kalınlığı (TVL), ortalama serbest yol (MFP), etkin atom numarası (Z_{etk}) ve etkin elektron yoğunluğu gibi çeşitli foton zırhlama parametreleri hesaplanabilmektedir.

Bu çalışmada saf PLA ve farklı oranlarda BaO içeren PLA/BaO kompozitlerinin kütle azaltma katsayıları (MAC)

XCOM kullanılarak elde edilmiştir. Elde edilen MAC değerleri kullanılarak LAC, HVL ve Z_{etk} değerleri hesaplanmıştır. Ayrıca, kompozitlerin MAC, LAC, HVL ve Z_{etk} değerleri Phy-X/PSD kullanılarak hesaplanmış ve XCOM değerleri ile karşılaştırılmıştır.

2.3. PLA/BaO Kompozitlerinin Kütle Azaltma ve Lineer Azaltma Katsayısı Değerleri

Saf PLA ve PLA/BaO kompozitlerinin kütle azaltma katsayıları XCOM programı kullanılarak 1 keV–100 GeV foton enerji aralığında hesaplanmıştır. Numuneler, saf PLA için PLA; ağırlıkça %5, %10 ve %20 BaO içeren kompozitler için sırasıyla PLA-5Ba, PLA-10Ba ve PLA-20Ba olarak adlandırılmıştır. Şekil 1’de, XCOM ile hesaplanan değerlerin 1 keV–1 MeV enerji aralığındaki değişimi gösterilmektedir.



Şekil 1. Saf PLA ve PLA/BaO kompozitlerinin kütle azaltma katsayılarının enerji ile değişimi

Şekil 1’den görüldüğü üzere, saf PLA ve PLA/BaO kompozitlerinin kütle azaltma katsayıları genel eğilim olarak

artan foton enerjisiyle azalmaktadır. Düşük enerji bölgesinde gözlenen yüksek kütle azaltma katsayıları, fotoelektrik etkileşimin bu enerji bölgesindeki baskınlığı ile ilişkilidir. Foton enerjisinin artmasıyla birlikte baskın etkileşim mekanizmasının değişmesi, kütle azaltma katsayısının enerjiye bağlı karakteristik davranışını belirlemektedir. Bu değişim literatürde bildirilen sonuçlarla uyumludur (Alshahri et al., 2021; El-Sayed Abdo et al., 2003; Prabhu, Bubbly, & Gudennavar, 2021). Bunun yanı sıra BaO katkı oranının artmasıyla kompozitlerin kütle azaltma katsayılarının arttığı görülmektedir. Daha yüksek kütle azaltma katsayısı değerleri, fotonların malzeme içerisinde daha etkin biçimde zayıflatıldığını göstermektedir. Dolayısıyla BaO ilavesinin PLA'nın foton zayıflatma performansını geliştirdiği söylenebilir. BaO içeren kompozitlerin eğrilerinde yaklaşık 37,44 keV'de gözlenen belirgin değişim, baryumun K-soğurma kenarından (K-edge) kaynaklanmaktadır.

PLA-20Ba kompozitinin kütle azaltma katsayıları, literatürde beton, HDPE/%30 PbO ve Epoksi/%20 nWO₃ için bildirilen değerlerle karşılaştırılmıştır (Alharshan et al., 2020; Gurler & Akar Tarım, 2012; Karabul & İçelli, 2021). 81 keV'de PLA-20Ba için hesaplanan 0,8277 cm²/g değeri, betonun 0,184 cm²/g ve HDPE/%30 PbO'nun 0,696 cm²/g değerlerinin sırasıyla yaklaşık 4,50 ve 1,19 katı olup, Epoksi/%20 nWO₃'ün 1,413 cm²/g değerinden düşüktür. 356 keV'de PLA-20Ba'nın MAC değeri 0,1128 cm²/g olarak hesaplanırken, beton, HDPE/%30 PbO ve Epoksi/%20 nWO₃ için bildirilen değerler sırasıyla 0,094, 0,156 ve 0,133 cm²/g'dır. 662 keV'de ise aynı sıralamayla bu değerler 0,08068, 0,075, 0,092 ve 0,089 cm²/g'dır. Böylece PLA-20Ba'nın MAC değerlerinin karşılaştırılan üç enerjide de betondan yüksek olduğu, HDPE/%30 PbO'ya göre üstünlüğünün ise yalnızca 81 keV'de görüldüğü belirlenmiştir.

Tablo 2’de belirli enerji değerleri için PLA ve PLA/BaO kompozitlerinin XCOM ve Phy-X/PSD ile hesaplanmış lineer azaltma katsayıları verilmektedir.

Tablo 2. PLA ve PLA/BaO kompozitlerinin lineer azaltma katsayıları

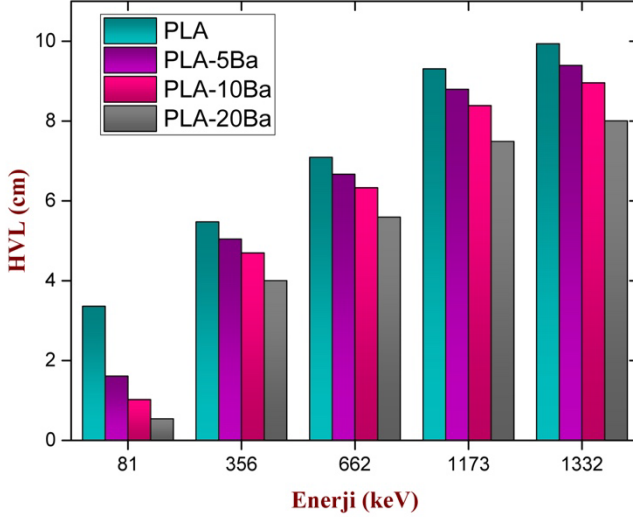
Örnek		Lineer Azaltma Katsayısı (cm^{-1})				
		81 keV	356 keV	662 keV	1173 keV	1332 keV
PLA	XCOM	0,206	0,127	0,098	0,074	0,070
	Phy-X/PSD	0,206	0,127	0,098	0,074	0,070
PLA-5Ba	XCOM	0,429	0,137	0,104	0,079	0,074
	Phy-X/PSD	0,429	0,137	0,104	0,079	0,074
PLA-10Ba	XCOM	0,675	0,148	0,109	0,083	0,077
	Phy-X/PSD	0,675	0,147	0,110	0,083	0,077
PLA-20Ba	XCOM	1,271	0,173	0,124	0,093	0,087
	Phy-X/PSD	1,270	0,173	0,124	0,093	0,087

Tablo 2’de görüldüğü üzere, saf PLA ve PLA/BaO kompozitleri için XCOM ve Phy-X/PSD kullanılarak hesaplanan lineer azaltma katsayıları birbiriyle oldukça uyumludur. Lineer azaltma katsayıları incelendiğinde, BaO katkı oranının artmasıyla μ değerlerinin sistematik olarak arttığı ve incelenen tüm enerji değerlerinde en yüksek değerlerin PLA-20Ba kompoziti için elde edildiği görülmektedir. Örneğin, 81 keV’de XCOM ile hesaplanan lineer azaltma katsayıları PLA-5Ba, PLA-10Ba ve PLA-20Ba için saf PLA değerinin sırasıyla yaklaşık 2,08, 3,28 ve 6,17 katıdır. Bu sonuçlar, BaO içeriğinin artırılmasının PLA’nın foton zayıflatma kabiliyetini belirgin biçimde geliştirdiğini ve incelenen kompozisyonlar arasında PLA-20Ba’nın en yüksek lineer azaltma performansına sahip olduğunu göstermektedir.

2.4. PLA/BaO Kompozitlerinin Yarı Değer Kalınlığı Değerleri

Saf PLA ve PLA/BaO kompozitlerinin yarı değer kalınlıkları (HVL), XCOM yazılımı ile hesaplanan kütle azaltma katsayılarından ve malzemelerin yoğunluk değerlerinden elde

edilen lineer azaltma katsayıları kullanılarak Denklem 3'e göre 1 keV–100 GeV foton enerji aralığında hesaplanmıştır. Şekil 2'de saf PLA ve PLA/BaO kompozitlerinin seçilen foton enerjilerindeki yarı değer kalınlıkları verilmektedir.



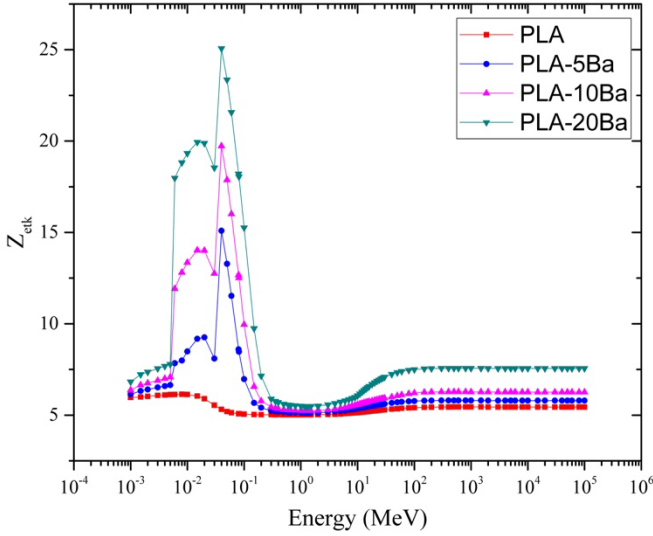
Şekil 2. PLA ve PLA/BaO kompozitlerinin belirli enerjilerdeki yarı değer kalınlığı değerleri

Şekil 2'de görüldüğü üzere, incelenen 81–1332 keV foton enerji aralığında saf PLA ve PLA/BaO kompozitlerinin yarı değer kalınlıkları (HVL), foton enerjisinin artmasıyla yükselirken BaO katkı oranının artmasıyla azalmaktadır. Yarı değer kalınlığı, bir zırhlayıcı malzemenin gelen foton şiddetini başlangıç değerinin yarısına düşürmesi için gerekli malzeme kalınlığı olarak tanımlanmaktadır. Bu nedenle yüksek HVL değerleri, fotonların malzeme içerisinde daha uzun bir mesafe kat ederek zayıflatıldığını ve dolayısıyla zırhlama performansının daha düşük olduğunu gösterirken, düşük HVL değerleri daha yüksek zırhlama performansına işaret etmektedir. Artan foton enerjisiyle birlikte tüm numunelerde HVL değerlerinin yükselmesi, foton enerjisi arttıkça malzemelerin zayıflatma etkinliğinin azaldığını göstermektedir. Bu davranış, artan foton enerjisiyle birlikte kütle

ve lineer azaltma katsayılarında gözlenen azalma ile uyumludur. Buna karşılık, BaO katkı oranının artmasıyla HVL değerlerinin azalması, BaO ilavesinin PLA'nın foton zayıflatma ve radyasyon zırlama performansını geliştirdiğini göstermektedir. İncelenen kompozitler arasında en düşük HVL değerlerinin PLA-20Ba numunesinde elde edilmesi, bu kompozitin en yüksek zayıflatma performansına sahip olduğunu ortaya koymaktadır. Sayısal olarak değerlendirildiğinde, 81 keV foton enerjisinde saf PLA için 3,36 cm olan HVL değeri, PLA-5Ba, PLA-10Ba ve PLA-20Ba kompozitleri için sırasıyla 1,61, 1,03 ve 0,55 cm'ye düşmüştür. Buna göre, saf PLA ile karşılaştırıldığında HVL değerinde PLA-5Ba, PLA-10Ba ve PLA-20Ba için sırasıyla yaklaşık %52,0, %69,5 ve %83,8 oranında azalma meydana gelmiştir. BaO katkısının HVL üzerindeki etkisinin düşük enerji bölgesinde daha belirgin olduğu görülmüş; PLA-20Ba için saf PLA'ya göre HVL değerindeki azalma oranı 1332 keV'de yaklaşık %19,4'e gerilemiştir.

2.5. PLA/BaO Kompozitlerinin Etkin Atom Numarası Değerleri

Saf PLA ve PLA/BaO kompozitlerinin etkin atom numaralarının (Z_{etk}) enerjiye bağlı değişimi, XCOM verileri ile Denklem 4 ve 5 kullanılarak hesaplanmış ve Şekil 3'te gösterilmiştir.



Şekil 3. PLA ve PLA/BaO kompozitlerinin etkin atom numarası-enerji grafikleri

Şekil 3'te saf PLA ve PLA/BaO kompozitlerinin etkin atom numaralarının (Z_{ek}) foton enerjisine bağlı değişimi gösterilmektedir. Eğrilerde görülen karakteristik değişim, literatürde bildirilen sonuçlarla uyumludur (Al-Buriahi et al., 2021; Çağlar et al., 2019; Prabhu et al., 2021). BaO içeren kompozitlerin Z_{ek} değerleri, özellikle düşük foton enerjilerinde saf PLA'ya göre belirgin biçimde yüksektir. Bu durum, fotoelektrik soğurmanın atom numarasına güçlü bağımlılığı ve kompozit yapı içerisindeki yüksek atom numaralı baryumun katkısı ile açıklanabilir. Yaklaşık 0,037 MeV civarında kompozitlerin eğrilerinde görülen keskin artış, baryumun K-kabuğu soğurma kenarı ile ilişkilidir.

Bu bölgenin ardından foton enerjisinin artmasıyla Z_{ek} değerleri hızla azalmakta ve Compton saçılmasının ağırlık kazandığı orta enerji bölgesinde eğriler birbirine yaklaşmaktadır. Daha yüksek enerjilerde ise çift oluşumunun artan katkısıyla Z_{ek} değerleri yeniden yükselmekte ve ardından yaklaşık sabit bir düzeye ulaşmaktadır. BaO katkı oranının Z_{ek} üzerindeki

etkisinin düşük enerji bölgesinde daha belirgin olduğu, orta enerji bölgesinde ise azaldığı görülmektedir.

İncelenen enerji aralığında en düşük Zetk değerleri saf PLA için elde edilirken, BaO katkı oranının artmasıyla bu değerler yükselmiş ve en yüksek değerler PLA-20Ba kompozitinde gözlenmiştir. Bu artış, kompozit yapı içerisindeki baryum oranının yükselmesiyle ilişkilendirilebilir. Zetk sonuçları, kütle azaltma katsayısı ve yarı değer kalınlığı sonuçlarıyla birlikte değerlendirildiğinde, incelenen bileşimler arasında PLA-20Ba kompozitinin teorik foton zırhlama özellikleri açısından daha elverişli olduğu görülmektedir.

3. SONUÇ VE ÖNERİLER

Bu çalışmada, ağırlıkça %5, %10 ve %20 BaO içeren PLA/BaO kompozitlerinin radyasyon zırhlama özellikleri XCOM ve Phy-X/PSD kullanılarak teorik olarak değerlendirilmiştir. Kompozitlerin foton zayıflatma davranışları; kütle azaltma katsayısı, lineer azaltma katsayısı, yarı değer kalınlığı ve etkin atom numarası üzerinden incelenmiştir.

Elde edilen sonuçlar, BaO ilavesinin PLA'nın radyasyon zayıflatma özelliklerini geliştirdiğini ve kütle azaltma katsayılarındaki katkı ile meydana gelen artışın fotoelektrik etkileşimin baskın olduğu düşük enerji bölgesinde daha belirgin olduğunu göstermiştir. İncelenen seçili enerjilerde BaO içeren kompozitlerin lineer azaltma katsayıları saf PLA'dan daha yüksek bulunmuş; özellikle 81 keV'de bu değerler saf PLA'nın yaklaşık 2,08–6,17 katı olarak hesaplanmıştır. BaO katkı oranının artmasıyla etkin atom numarası yükselmiş, yarı değer kalınlığı ise azalmıştır. İncelenen kompozisyonlar arasında PLA-20Ba, seçili enerjilerde en yüksek lineer azaltma katsayısına ve en düşük yarı değer kalınlığına sahip olmuştur.

Literatür karşılaştırmasında, PLA-20Ba'nın 81 keV'deki kütle azaltma katsayısı betonun yaklaşık 4,50 katı ve HDPE/%30 PbO kompozitinden %18,9 daha yüksek bulunmuştur. Bununla birlikte, 356 ve 662 keV'de bu değerler betondan yüksek, HDPE/%30 PbO ve Epoksi/%20 nWO₃ kompozitlerinden düşük kalmıştır. Bu sonuçlar, PLA-20Ba'nın özellikle düşük enerjili fotonların zayıflatılması açısından kurşun içermeyen polimer esaslı zırhlama malzemeleri arasında potansiyel bir aday olabileceğini göstermektedir. Uygulama potansiyelinin belirlenmesi için teorik sonuçların deneysel ölçümlerle desteklenmesi ve kompozitlerin mekanik ve termal özelliklerinin incelenmesi önerilmektedir.

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BORON-HYDRIDE SYSTEMS: SUPERCONDUCTIVITY UNDER HIGH PRESSURE AND THE QUEST FOR ROOM- TEMPERATURE SUPERCONDUCTIVITY

Özge ERDEM¹

1. INTRODUCTION

One of the fundamental goals in superconductivity research is to achieve the phase, characterized by zero electrical resistivity and the Meissner effect, stably at the highest possible critical temperatures (T_c). To this end, experimental and theoretical studies conducted on hydride-based systems under high pressure have shown that T_c values can approach room temperature due to enhanced phonon-mediated pairing mechanisms. The transition of hydrogen-rich compounds into a metallic phase under high pressure, together with their high-frequency lattice vibrations and strong electron–phonon coupling, suggests that exceptionally high critical temperatures may be possible within the framework of BCS theory. T_c values measured above 200 K in some super hydride phases demonstrate the critical importance of these systems for high-temperature superconductivity. These studies have brought the concept of “room-temperature superconductivity” closer to becoming an experimentally accessible limit rather than remaining merely a theoretical goal.

Within the super hydride class, boron-based hydrides constitute a prominent area of research due to the combination of

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the high-frequency phonon spectra generated by low-atomic-mass elements and the versatile covalent bonding chemistry of boron. The electron-deficient nature of boron enables the formation of multicentre bonds and complex lattice structures, while these characteristics make the emergence of new superconducting phases possible under high pressure. In boron–hydride compounds, the complex crystalline networks stabilized under high pressure, together with the accompanying strong electron–phonon interactions, provide a favourable basis for achieving high T_c within the framework of BCS theory. Furthermore, the structural phase diversity exhibited by these systems enables the engineering of their electronic band structures and phonon densities of states, thereby supporting the development of new design strategies for superconductivity at temperatures approaching room temperature. However, the fact that a large fraction of these phases are thermodynamically stable only under pressures on the order of megabars (Mbar, 106 bar) represents a significant limitation for practical applications. Therefore, current research focuses, on the one hand, on the discovery of boron–hydride phases that are stable at lower pressures and, on the other hand, on the kinetic trapping of structures synthesized under high pressure to preserve them under ambient conditions.

This section discusses boron–hydride superconductors and research efforts toward achieving room-temperature superconductivity.

2. SUPERCONDUCTIVITY IN BORON–HYDRIDE COMPOUNDS

2.1. Phonon-Mediated Superconductivity

In boron–hydride systems, according to the conventional mechanism of superconductivity, superconductivity is explained

within the framework of BCS theory by the formation of Cooper pairs through phonon-mediated electron pairing. When strong interactions occur between the Cooper pairs, the Migdal–Eliashberg theory, which is a generalization of BCS theory, is employed.

The critical temperature of superconductors depends on the electron–phonon coupling constant (λ), the average phonon frequency, and the electronic density of states. The electron–phonon coupling constant is a dimensionless parameter in condensed-matter physics, particularly in theories of superconductivity and metallic conduction, that characterizes the strength of the interaction between electrons and lattice vibrations (phonons); in other words, it indicates how strongly electrons interact with the crystal lattice. Electrical resistance arises from the scattering of electrons by phonons, and the scattering rate depends on the value of λ . The larger the value of λ , the stronger the electron–phonon interaction responsible for the formation of Cooper pairs in BCS theory. For materials such as high-temperature superconductors, $\lambda > 1$ represents strong coupling and is generally associated with a high T_c value [1, 2].

In superconductors, the phonon frequency is the characteristic frequency of the crystal lattice vibrations (phonons) that enable electrons to come together and form Cooper pairs, particularly within the framework of conventional (BCS) superconductivity theory. The phonon frequency in superconductors varies depending on the crystal structure and atomic mass of the material, and the characteristic phonon frequencies play an important role in determining the electron–phonon interaction and, consequently, the superconducting transition temperature [2–4]. The Debye frequency (ω_D), on the other hand, refers to the cutoff frequency that limits the phonon spectrum in the Debye model and determines the total number of vibrational modes in the crystal [5]. In particular, within the

framework of Migdal–Eliashberg theory, the transition temperature can be calculated using the electron–phonon coupling parameter and the logarithmic average phonon frequency [1, 2]. During the transition to the superconducting state, the amplitude of lattice vibrations decreases as the temperature is lowered; however, the specific frequency and energy interactions required for the transition to the superconducting state, namely the electron–phonon interaction responsible for Cooper pair formation, persist [4, 6]. Light elements such as hydrogen (H) and boron (B) provide an advantage for achieving high T_c values due to their high phonon frequencies.

2.2. Effect of High Pressure

High pressure can reduce the interatomic distances, leading to the broadening and overlap of electronic bands; this can facilitate the emergence of a metallic state in some hydrogen-rich compounds [7]. Metallization is important for superconductivity because electrons must be mobile, and since free electrons are present near the Fermi level only in the metallic phase, the electron–phonon interaction becomes effective, enabling the formation of Cooper pairs [4, 6, 8].

In hydrogen-containing systems, high pressure can alter the stability of molecular structures, leading to the formation of hydrogen-rich metallic phases. In such metallic phases, high-frequency hydrogen vibrations and strong electron–phonon interactions can create favourable conditions for conventional superconductivity with high T_c values [9, 10]. During metallization in boron–hydride systems, increasing pressure brings the atoms closer together, enhances orbital overlap, increases band widths, and eventually closes the band gap, resulting in the formation of free charge carriers [5, 7, 11].

2.3. Hydride Superconductors

Although the majority of hydride systems do not exhibit superconductivity at ambient pressure, high pressure can enable the formation of new crystalline phases and the emergence of metallic and superconducting states. In particular, super hydride phases such as LaH_{10} have been synthesized at pressures on the megabar scale, with high T_c values of up to 260 K being reported [7, 10, 12]. Hydrogen-rich metal hydrides are considered promising systems for reducing the external pressure required to metallize pure hydrogen through chemical pre-compression, while atomic metallic hydrogen itself has also been theoretically investigated as a potential high T_c superconductor under high pressure [7, 13]. In such compounds, high pressure can induce metallization by narrowing the electronic band gap and eventually closing it above a certain pressure. With the formation of the metallic phase, a finite and high density of states emerges near the Fermi level, providing the necessary condition for electronic conductivity [14]. In the metallic phase, strong electron–phonon interactions between the electrons and lattice vibrations can contribute to the formation of Cooper pairs and the emergence of conventional superconductivity [14, 15]. As a result of this pairing mechanism, the system transitions into the superconducting phase, exhibiting zero electrical resistivity and magnetic-field expulsion characterized by the Meissner effect.

In hydride systems, the low atomic mass of hydrogen leads to high-frequency phonon modes and, consequently, a high Debye frequency [7, 14, 16]. Together with a large electron–phonon coupling constant (λ) and a suitable electronic band structure, these characteristics enable high T_c values, as described within the framework of BCS theory and its strong-coupling generalization, the Eliashberg formalism [15, 17].

Hydrogen-rich metal hydrides such as H_3S (~ 200 K) [18], LaH_{10} (~ 250 K) [19], and YH_6 (~ 220 K) [20] are among the most prominent systems in the field of high-temperature superconductivity. These compounds, referred to as “super hydrides” due to their high hydrogen content, can exhibit T_c values approaching room temperature; however, they face significant limitations for technological applications because they generally become stable only under extremely high pressures, well above 100 GPa and in the mega bar range [12, 19, 21].

2.4. Boron–Hydride Superconductors

Boron is an electron-deficient element and, in its stable compounds, exhibits multicentre bonding, such as three-centre two-electron bonds, rather than conventional two-centre bonding. This bonding character enables boron to form complex and highly interconnected lattice structures. In boron–hydride systems, these characteristics result in enhanced structural stability, the emergence of high-frequency phonon modes, and the formation of a wide variety of phases. In this context, boron–hydride compounds can generally be classified as simple boron hydrides, metal-doped boron hydrides, and boron–carbon–hydride systems [22, 23].

Simple boron hydrides include low-atom-number molecular structures such as BH_3 , B_2H_6 , and B_4H_{10} . These compounds are among the most basic hydrides formed by boron and hydrogen. They generally exhibit well-defined molecular structures but limited thermodynamic stability under ambient conditions due to their electron-deficient bonding [24, 25]. Because these molecular systems exhibit insulating or wide-band-gap semiconducting characteristics in terms of their electronic band structures, they do not exhibit superconductivity [17, 25]. However, under high pressure, these molecular phases can transform into denser, periodic crystalline structures, leading to

band-gap closure and metallization. This transformation enables the electron–phonon interaction mechanism to become effective, thereby creating favourable conditions for the formation of superconducting phases. Nevertheless, pure boron–hydride systems generally cannot provide the electron–phonon coupling required to achieve high T_c values. Therefore, high T_c superconductivity is predominantly observed in metal-doped or more complex super hydride systems.

Compounds such as LiBH_4 , NaBH_4 , and $\text{Mg}(\text{BH}_4)_2$ belong to the class of metal-doped boron–hydride systems and have been widely investigated in hydrogen-storage studies [26, 27]. LaBH_8 , on the other hand, is an example of a ternary super hydride phase that is stable under high pressure. In such ternary hydrides, the chemical structure formed by the combination of different elements can modify the stability and electronic properties of the hydrogen sublattice. In the specific case of LaBH_8 , the La–B framework may stabilize the metallic character of the hydrogen sublattice through a chemical-pressure effect, thereby creating favourable conditions for superconductivity [28, 29]. The metal atom alters the electronic structure of the boron–hydrogen sublattice by providing electrons through charge transfer; this can contribute to narrowing the band gap and facilitating the transition to the metallic state. At the same time, the incorporation of the metal can enhance the thermodynamic stability of the crystal structure and enable the stabilization of different high-pressure phases.

From an electronic-structure perspective, metal doping increases the density of states near the Fermi level, creating favourable conditions for strong electron–phonon interactions. In these systems, the high-frequency phonon modes associated with the low atomic mass of hydrogen, together with the enhanced electron–phonon interaction, support superconducting pairing

within the framework of BCS theory and the Eliashberg formalism [15, 17, 28, 29].

Some metal-doped boron–hydride phases are theoretically predicted to reach critical temperatures above 100 K at pressures of approximately 50–100 GPa. In particular, theoretical studies of boron–hydride systems containing rare-earth and alkali/alkaline-earth metals, such as the La–B–H and Mg–B–H phase diagrams, support this range [28, 29]. Although such ternary boron–hydride systems may exhibit lower critical temperatures than super hydrides, they constitute an important area of research for the design of practical superconducting materials due to their potential to remain stable at lower pressures.

Boron–carbon–hydride (B–C–H) systems are compounds formed by the hydrogenation of the covalent framework composed of boron and carbon atoms, exhibiting rich structural and electronic properties [21, 29]. The combination of boron’s electron-deficient bonding character and carbon’s tendency to form four-valent covalent bonds enables the formation of complex structures with greater thermodynamic and kinetic stability compared with conventional boranes. Under ambient conditions, these systems generally exhibit insulating or semiconducting electronic band structures, while under high pressure, they can transition to a metallic phase through the closure of the band gap.

Carbon incorporation can modify the electron distribution within the boron–hydrogen sublattice, thereby tuning the electronic states near the Fermi level, contributing to the adjustment of the band structure, and improving the structural stability of certain high-pressure phases [30–32]. Structurally, boron and carbon atoms in these compounds can form polyhedral clusters and lattice-like cluster structures; while multicentre bonding motifs are preserved, a transformation into extended

three-dimensional covalent crystal networks may occur under high pressure. Such structural changes directly influence the reconstruction of the electronic band structure and the metallization process.

B–C systems offer potential candidates for high-temperature superconductivity by taking advantage of high-frequency vibrational modes arising from their covalent lattice structures composed of light elements. Theoretical studies have shown that high T_c values can be achieved in metal-doped B–C clathrates through the tuning of the electronic density of states and lattice dynamics [30]. Furthermore, incorporating hydride units into B–C clathrate structures provides a design approach for enhancing electron–phonon interactions and, consequently, superconducting properties by exploiting the low atomic mass of hydrogen [31]. For example, T_c values on the order of 100 K at ambient pressure have been theoretically predicted for some B–C–H-based clathrates [30, 31].

B–C–H compounds are being extensively investigated not only in the context of superconductivity but also in areas such as high-hardness materials [33], nanostructures, hydrogen-storage technologies [22], and energy materials. This versatility further enhances the importance of these systems in both fundamental science and applied materials science.

The synthesis of the vast majority of boron–hydride superconductors exhibiting high T_c requires extremely high pressures in the megabar range (≥ 100 GPa), while the resulting phases are generally limited to micron-scale samples. This constitutes a fundamental limitation in terms of the scalability and technological applicability of these materials. Furthermore, most of these phases become thermodynamically unstable when the pressure is released and cannot be preserved under ambient conditions.

In this context, current research focuses on material-design strategies aimed at overcoming these limitations. In particular, element substitution (chemical doping) is being explored to tune lattice parameters and the electronic structure, thereby reducing the external pressure required to stabilize the desired phases [29, 34]. In addition, kinetically trapping phases obtained under high pressure through pressure-induced metastability or controlled decompression pathways, thereby preserving metastable structures at lower pressures or under conditions close to ambient pressure, has emerged as an important area of research [21, 29]. On the other hand, in the context of low-dimensional systems, two-dimensional boron derivatives offer tunable electronic platforms due to their diverse crystal structures and the resulting variation in their electronic properties [35, 36]. Functionalization of these structures with hydrogen or other elements may provide potential superconducting platforms by enabling control over the electronic structure and electron-phonon interactions. These approaches represent key research directions aimed at achieving high T_c values within the framework of BCS theory under more accessible pressure and temperature conditions.

3. CONCLUSIONS

Boron-hydride systems constitute an important research platform in condensed-matter physics in the context of phonon-mediated superconductivity observed under high pressure. The combination of the high T_c potential of hydrogen-rich hydrides with the structural flexibility of boron based on multicentre bonding makes the goal of achieving superconductivity at temperatures approaching room temperature theoretically accessible, provided that suitable electronic band structures and strong electron-phonon interactions are achieved. Within this

framework, the mechanism is largely based on high-frequency phonon modes, which are described by BCS theory and the Eliashberg formalism.

However, the fact that the vast majority of these systems can only be stabilized under pressures in the megabar range constitutes the most fundamental obstacle to their transition to practical applications. Therefore, current research focuses on reducing the required external pressure through approaches such as chemical pre-compression and element substitution (chemical doping), as well as transforming phases obtained under high pressure into metastable structures that can be preserved under ambient conditions. Such strategies are considered critical steps toward achieving high T_c values under more accessible thermodynamic conditions.

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TOPRAKTAN SOFRAYA DOĞAL RADYONÜKLİTLER: GIDA ZİNCİRİNDE TRANSFER, İÇ DOZ VE RADYOLOJİK RİSK

Gülçin Bilgici CENGİZ¹

1. GİRİŞ

İnsanlar doğal ve insan kaynaklı iyonlaştırıcı radyasyona yaşamları boyunca maruz kalmaktadır. Doğal maruziyetin önemli bir bölümü; yerkabuğu, toprak, su, hava ve canlı dokularda bulunan radyonüklitlerden kaynaklanır. Uranyum ve toryum bozunma serilerinin üyeleri ile doğal potasyumun radyoaktif izotopu olan ⁴⁰K, gıda kaynaklı radyolojik değerlendirmelerde öne çıkmaktadır (UNSCEAR, 2000).

Toprakтан bitkilere geçen radyonüklitler, bitkisel ürünlerin doğrudan tüketilmesi veya yem–hayvan–hayvansal ürün zinciri üzerinden insanlara ulaşabilir. Kök alımına ek olarak atmosferik çökeltme ve yüzeyde tutunan toprak parçacıkları da bitkisel ürünlerde ölçülen aktiviteye katkıda bulunabilir (IAEA, 2010).

Gıdalardaki aktivite konsantrasyonlarının sağlık açısından anlamı; tüketim miktarı, tüketici yaşı ve radyonüklide özgü yutma dozu katsayılarıyla birlikte değerlendirilir (ICRP, 2012). Bu nedenle yüksek aktivite içeren fakat az tüketilen bir ürünün doz katkısı, daha düşük aktiviteye sahip ancak yaygın tüketilen temel bir gıdadan az olabilir.

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Bu bölümde doğal radyonüklitlerin topraktaki davranışı, bitkisel ve hayvansal gıdalara transferi, işleme süreçlerindeki değişimi ve tüketim yoluyla oluşan iç doz ile radyolojik risk bütüncül olarak ele alınmaktadır.



Şekil 1. Doğal radyonüklitlerin topraktan sofraya başlıca transfer yolları.

2. DOĞAL RADYONÜKLİTLERİN TOPRAKTAKİ DAVRANIŞI

Toprak, karasal ekosistemlerde doğal radyonüklitlerin temel kaynak ve birikim ortamıdır. Aktivite düzeyleri jeolojik yapı, ana materyal, iklim ve toprak oluşum süreçlerine; hareketlilikleri ise toprağın fiziksel ve kimyasal özelliklerine bağlıdır.

2.1. Doğal Radyonüklitlerin Çevresel Kaynakları

Doğal radyonüklitler, yerkürenin oluşumundan beri çevrede bulunan birincil radyonüklitler ile bunların bozunma ürünlerinden oluşmaktadır. Gıda zincirine transfer bakımından en fazla incelenen doğal radyonüklitler, ^{238}U ve ^{232}Th bozunma serilerinin üyeleri ile ^{40}K 'dir. Bu radyonüklitler kayaçların ayrışması, erozyon, yüzeysel akış ve sediment taşınımı gibi doğal süreçlerle toprağa geçmekte ve toprak profilinin farklı katmanlarında birikebilmektedir (UNSCEAR, 2000).

Tablo 1. Gıda zinciri açısından önemli doğal radyonüklitlerin temel özellikleri ve çevresel davranışları

Radyonüklit	Yarı ömür	Başlıca çevresel kaynak	Toprak ve bitkilerdeki davranışı	Gıda zinciri açısından önemi
^{226}Ra	Yaklaşık 1600 yıl	^{238}U bozunma serisi; kayalar ve toprak	Kalsiyuma kimyasal benzerlik gösterir. Toprak özelliklerine bağlı olarak kökler tarafından alınabilir; bitkilerdeki transferi genellikle sınırlı ve değişkendir.	Bitkisel ürünler, yem, süt ve diğer hayvansal ürünler yoluyla alınabilir. Vücuda geçtiğinde özellikle kemik dokusuyla etkileşebilir.
^{232}Th	Yaklaşık $1,4 \times 10^{10}$ yıl	Toryum içeren kayalar, mineraller ve bunlardan oluşan topraklar	Kil mineralleri, metal oksitler ve organik maddelere güçlü biçimde bağlanır. Hareketliliği ve kök yoluyla bitkilere transferi genellikle düşüktür.	Gıdalarda çoğunlukla düşük düzeylerde bulunur. Bitkilerde ölçülen aktiviteye yüzeye tutunan toprak ve toz parçacıkları katkıda bulunabilir.
^{40}K	Yaklaşık $1,25 \times 10^9$ yıl	Doğal potasyum; toprak mineralleri ve potasyum içeren gübreler	Potasyum temel bir bitki besin elementidir. Kökler tarafından aktif olarak alınır ve bitki dokularına taşınır.	Bitkisel ve hayvansal gıdalarda yaygın olarak bulunur. Vücuttaki potasyum miktarı fizyolojik mekanizmalarla düzenlenir.

Topraklardaki doğal radyonüklit düzeylerinin temel belirleyicisi jeolojik yapıdır. Granitik ve fosfatça zengin kayalardan gelişen topraklar, bazı doğal radyonüklitler bakımından daha yüksek aktivite gösterebilirken farklı ana materyaller üzerinde oluşan topraklarda daha düşük değerler belirlenebilir. Kayaların fiziksel ve kimyasal ayrışmasıyla serbest kalan radyonüklitler; toprak minerallerine bağlanabilir,

toprak çözeltisine geçebilir, erozyonla taşınabilir veya bitki kökleri tarafından alınabilir. Bu nedenle aynı bölge içerisinde dahi farklı toprak türleri ve arazi kullanım biçimleri arasında önemli değişimler görülebilir.

Doğal süreçlerin yanı sıra bazı insan faaliyetleri de doğal radyonüklitlerin çevresel dağılımını değiştirebilir. Özellikle fosfatlı gübrelerin uzun süreli ve yoğun kullanımı, tarım topraklarına uranyum ve radyum serisi radyonüklitlerin eklenmesine neden olabilir (Alam ve ark., 1997; Boukhenfouf ve Boucenna, 2011; Ghosh ve ark., 2008; Hussain ve Hussain, 2011).

Madencilik, kömür yakılması, petrol ve doğal gaz üretimi, cevher işleme ve bazı endüstriyel faaliyetler sırasında doğal olarak bulunan radyoaktif maddeler çevresel ortamlarda yoğunlaşabilir. Bu maddeler, teknolojik olarak zenginleştirilmiş doğal radyoaktif maddeler olarak değerlendirilmektedir. Ancak bu faaliyetlerin tarım toprakları üzerindeki etkisi; kullanılan hammaddenin özelliklerine, uygulama süresine, bölgesel koşullara ve çevresel kontrol önlemlerine bağlıdır (IAEA, 2003).

2.2. Toprak Özellikleri, Hareketlilik ve Biyoyararlanabilirlik

Toprağın pH'sı, kil ve organik madde içeriği, katyon değişim kapasitesi, nem ve redoks koşulları radyonüklitlerin çözünürlüğünü, tutulmasını ve bitkiler tarafından alınabilirliğini belirler. Asidik koşullar bazı radyonüklitlerin hareketliliğini artırabilirken güçlü adsorpsiyon transferi sınırlandırabilir (Rachkova ve ark., 2010). Toplam aktivite konsantrasyonu bitkiye geçişi doğrudan yansıtmaz; alım, çoğunlukla toprak çözeltisindeki ve zayıf bağlanmış fraksiyonlardan gerçekleşir. Hareketlilik yüksek olsa bile kimyasal form, diğer elementlerle rekabet ve köklerin seçiciliği biyoyararlanabilirliği sınırlayabilir (IAEA, 2010).

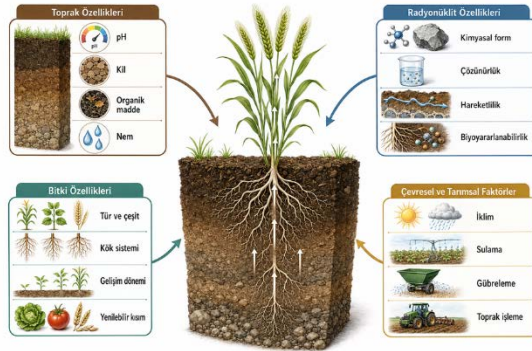
3. TOPRAKTAN BİTKİLERE RADYONÜKLİT TRANSFERİ

Topraktan bitkilere radyonüklit transferi, gıda zinciri yoluyla iç maruziyetin oluşmasında önemli bir aşamadır. Transferin değerlendirilmesinde radyonüklitlerin bitkiye ulaşma yolları, bitki içindeki dağılımı ve yenilebilir kısımlardaki aktivite konsantrasyonları dikkate alınır.

3.1. Transfer Yolları ve Etkileyen Faktörler

Radyonüklitler bitkilere kök alımı, atmosferik çökeltme ve toprak parçacıklarının yüzeye tutunması yoluyla ulaşır. Köklerle alınan radyonüklitlerin bir bölümü köklerde tutulurken diğer bölümü gövde, yaprak ve tanelere taşınır. Havadaki aerosol ve tozlar kuru veya yaş çökeltmeyle; toprak parçacıkları ise rüzgâr, yağmur sıçraması, sulama ve hasat sırasında bitki yüzeylerine ulaşabilir (IAEA, 2010).

Transfer; toprak özellikleri, radyonüklidin kimyasal formu, bitki türü, gelişim dönemi ve kök yapısı ile iklim, sulama ve gübrelemenin ortak etkisine bağlıdır (Şekil 2). Aynı toprakta yetişen bitkiler farklı transfer değerleri gösterebilir. Bitki içindeki dağılım da homojen olmadığından radyolojik değerlendirmelerde doğrudan tüketilen kısmın analizi önemlidir (IAEA, 2010).



Şekil 2. Doğal radyonüklitlerin topraktan bitkilere transferini etkileyen başlıca faktörler.

3.2. Transfer Faktörü ve Tahıl-Un Zinciri

Toprak-bitki transfer faktörü aşağıdaki oranla tanımlanır:

$$TF = \frac{A_{bitki}}{A_{toprak}}$$

Burada A_{bitki} ve A_{toprak} , aynı radyonüklidin bitki ve topraktaki, aynı kütle esasına göre verilen aktivite konsantrasyonlarını ($Bq\ kg^{-1}$) ifade eder. Örnekler aynı yer ve dönemde alınmalı, yüzeysel kontaminasyon kontrol edilmeli ve incelenen bitki kısmı belirtilmelidir. Transfer faktörleri evrensel sabitler değil, belirli toprak-bitki-iklim koşullarının göstergeleridir (IAEA, 2010).

Tahıllar yüksek tüketimleri nedeniyle radyolojik açıdan önemlidir. Doğal radyonüklitlerin buğdayın kök, gövde ve tane kısımlarındaki dağılımı farklılık gösterebilir (Pulhani ve ark., 2005). Öğütmenin ürün fraksiyonlarındaki aktiviteyi değiştirebildiği, radyoaktif sezyumla yapılan çalışmalarda da gösterilmiştir (Kimura ve ark., 2012). Bu nedenle toprak-buğday ve toprak-un oranları ayrı değerlendirilmelidir. Toprak-un oranı, işleme etkilerini de içeren bütünsel bir toprak-ürün göstergesidir.

4. BİTKİSEL VE HAYVANSAL GIDALARDA DOĞAL RADYONÜKLİTLER

Doğal radyonüklitler bitkisel ürünlerin doğrudan tüketimi veya yem-hayvan zinciri üzerinden gıdalara ulaşır. Nihai düzeyler ürün türü, yetiştirme ortamı, işleme ve hayvan metabolizması gibi çok sayıda faktöre bağlıdır.

4.1. Tahıllar ve Tahıl Bazlı Ürünler

Buğday, arpa, yulaf, mısır ve pirinç ile bunlardan üretilen un, ekmek, makarna, bulgur ve bisküvi gibi ürünler, yaygın

tüketimleri nedeniyle toplam iç doza katkı sağlayabilir (Bilgici Cengiz ve ark., 2025). İşlenmiş ürünlerdeki aktivite; ham madde, ürün bileşimi, pişirme ve nem oranından etkilenir. Bu nedenle değerlendirmelerde tüketime hazır ürünün aktivitesi ve tüketim miktarı birlikte dikkate alınmalıdır (IAEA, 2010).

4.2. Sebzeler ve Geleneksel Kurutulmuş Sebze Ürünleri

Sebzelerdeki aktivite, tüketilen bitki kısmı ve yüzeysel çökelmeden etkilenir (IAEA, 2010). Kurutma, su kaybıyla birim kütle başına aktiviteyi artırabilir; açık havada kurutma ise toz ve toprak parçacıklarının yüzeye tutunmasına yol açabilir. Bu etkileri ayırt etmek için işlem öncesi ve sonrası sonuçlar aynı kuru madde esasına göre karşılaştırılmalıdır (IAEA, 1992).

4.3. Tıbbi-Aromatik Bitkiler ve Baharatlar

Tıbbi-aromatik bitki ve baharatlarda aktivite, kullanılan bitki kısmı, yetiştirme ortamı ve yüzeyde tutunan tozdan etkilenir. Düşük tüketim miktarları yıllık doz katkısını sınırlayabilir; yoğun ve uzun süreli kullanım ise ayrıca değerlendirilmelidir. Bitkisel çaylarda doz hesabı, kuru ürünlerdeki toplam aktivite yerine demleme sırasında suya geçen fraksiyona dayanmalıdır (Grządka ve ark., 2024).

4.4. Hayvansal Gıdalar ve Süt Ürünleri

Hayvanlar radyonüklitleri yem, su, mineral takviyeleri ve otlama sırasında yutulan toprak yoluyla alır. Et ve süte transfer, radyonüklidin kimyasal özellikleri, beslenme ve hayvan metabolizmasına bağlıdır (IAEA, 2010). Sütün işlenmesi sırasında radyonüklitler peynir ve peynir altı suyu arasında farklı oranlarda dağılır; su kaybı da nihai aktivite konsantrasyonunu etkiler (IAEA, 2010). Transferin değerlendirilmesi için toprak, yem, süt ve hayvansal ürünlerin eş zamanlı incelenmesi önemlidir.

Türkiye’de ve farklı ülkelerde gerçekleştirilen çalışmalarda çeşitli gıda grupları için bildirilen doğal radyonüklit aktivite konsantrasyonları ile yıllık etkin yutma dozlarının seçilmiş örnekleri Tablo 2’de karşılaştırılmıştır.

Tablo 2. Türkiye ve farklı ülkelerde çeşitli gıda grupları için bildirilen doğal radyonüklit aktivite konsantrasyonları ve yıllık etkin yutma dozları

Ülke/bölge	Gıda grubu	²²⁶ Ra (Bq kg ⁻¹)	²³² Th (Bq kg ⁻¹)	⁴⁰ K (Bq kg ⁻¹)	Yıllık etkin yutma dozu (μSv yıl ⁻¹)	Kaynak
Türkiye, Kars	Tahıl, et, peynir, fırın ürünleri ve kaya tuzu	2,20– 36,20	2,98– 49,56	83,55– 304,89	18,37–195,84 ¹	Bilgici Cengiz ve Çağlar (2022)
Türkiye	Kahvaltılık tahıllar	12,59 ± 1,19	10,64 ± 0,80	87,08 ± 14,71	143; 130 ²	Bilgici Cengiz ve ark. (2025)
Türkiye	Bisküviler	12,42 ± 1,15	10,48 ± 1,11	77,54 ± 14,67	140; 127 ²	Bilgici Cengiz ve ark. (2025)
Türkiye, Güneydoğu Anadolu	Baharatlar	25,70 ± 2,12	24,43 ± 1,65	409,67 ± 24,10	27,64 ³	Cengiz ve ark. (2026)
Türkiye	Süt ve süt ürünleri	<MDA– 16,0	<MDA– 9,8 ⁴	20,6– 658,4	H	Ağuş (2017)
Türkiye, Elazığ	Sebzeler	0,64 ± 0,26	0,65 ± 0,14	13,98 ± 1,22	20,00	Canbazoglu ve Dogru (2013)
Türkiye, Elazığ	Meyveler	1,52 ± 0,34	0,98 ± 0,23	18,66 ± 1,13	30,55	Canbazoglu ve Dogru (2013)
Malezya	Buğday, yulaf ve arpa	56,97– 70,52	34,71– 49,57	517,05– 995,90	H	Salih (2018)
Suudi Arabistan	Buğday ve arpa	3,91– 10,63	3,20– 6,33	139,26– 178,31	348,83 ⁵	Baz ve Alamoudi (2017)
Pakistan	Buğday	1,40 ± 0,30	1,30 ± 0,20	122,8 ± 23,1	216,9	Tufail ve ark. (2010)
Suudi Arabistan	Siğır ve tavuk eti	3,30– 6,37	2,48– 6,49	104,86– 166,47	150,60 ⁵	Baz ve Alamoudi (2017)
Mısır	Peynir	–	–	58	H	Gomaa ve ark. (1995)
Irak, Basra	Makarna	0,72 ± 0,39	0,97 ± 0,99	209,17 ± 71,12	H	Alsalihi ve ark. (2017)
Ürdün	Tuz	2 ± 1	9 ± 5	82 ± 57	H	Shaltout ve ark. (2017)

Not: Aktivite konsantrasyonları, ilgili çalışmalarda bildirilen ortalama ± belirsizlik veya minimum–maksimum değerleridir. Karşılaştırmalarda çalışmaların numune hazırlama yöntemleri ve yaş veya kuru kütle esasları dikkate alınmalıdır.

¹ İncelenen tekil gıdalar için bildirilen yıllık etkin yutma dozu aralığıdır. Çalışmada incelenen bütün gıdaların tüketimine bağlı toplam yıllık etkin yutma dozu 984,32 μSv yıl⁻¹ olarak bildirilmiştir.

² İlk değer çocuklar, ikinci değer yetişkinler için hesaplanan yıllık etkin yutma dozudur.

³ Yetişkin tüketim senaryosu için bildirilen ortalama yıllık etkin yutma dozudur.

⁴ ²³²Th, yalnızca süt tozu örneklerinde 3,2–9,8 Bq kg⁻¹ aralığında belirlenmiş; diğer süt ve süt ürünlerinde MDA’nın altında kalmıştır.

⁵ Yıllık etkin doz, ilgili çalışmada aynı gıda grubunun genel tüketim senaryosu için bildirilmiştir.

“H”, yıllık etkin yutma dozunun hesaplanmadığını; “–”, ilgili radyonüklidin raporlanmadığını; “<MDA” ise aktivitenin

minimum dedekte edilebilir aktivitenin altında olduğunu göstermektedir.

Tablo 2'deki farklılıklar; çevresel koşullar, gıda türü, transfer ve işleme süreçleri ile ölçüm yöntemlerinden kaynaklanabilir. Dozlar ayrıca tüketim miktarı, yaş ve yutma dozu katsayılarına bağlı olduğundan çalışmalar karşılaştırılırken bu değişkenler ve kütle esas dikkate alınmalıdır (IAEA, 2010; ICRP, 2012).

5. GIDA İŞLEME SÜREÇLERİNİN RADYONÜKLİT DÜZEYLERİNE ETKİSİ

Yıkama, kabuk soyma, öğütme, kurutma, kaynatma ve sütü peynire dönüştürme gibi işlemler radyonüklitleri uzaklaştırabilir, yoğunlaştırabilir veya ürün fraksiyonları arasında yeniden dağıtabilir. Etkinin yönü radyonüklidin kimyasal formu, işlem koşulları ve kütle değişimine bağlıdır (Green ve Wilkins, 1996; IAEA, 2010).

İşlem öncesi ve sonrası aktivite konsantrasyonları tek başına yeterli değildir; kuru madde oranı, ürün verimi ve toplam aktivite dengesi de dikkate alınmalıdır (IAEA, 2010).

5.1. Yıkama, Kabuk Ayırma ve Öğütme

Yıkama yüzeye tutunan toz ve toprak parçacıklarını, kabuk ayırma ise dış tabakalardaki aktivitenin bir bölümünü uzaklaştırabilir. Öğütme ve eleme, radyonüklitlerin kepek, ruşeym ve un fraksiyonları arasındaki dağılımını değiştirir. İşlemlerin etkisi değerlendirilirken ürün verimi ve kütle kaybı dikkate alınmalıdır (IAEA, 2010; Green ve Wilkins, 1996).

5.2. Kurutma, Kaynatma ve Pişirme

Kurutma, fırınlama ve kavurma sırasında su kaybı, toplam aktivite değişmese bile birim kütle başına aktiviteyi artırabilir.

Radyonüklit kaybı veya dışarıdan bulaşma olmadığında kurutma etkisi şöyle ifade edilir:

$$A_{kuru} = A_{yaş} \times \left(\frac{m_{yaş}}{m_{kuru}} \right)$$

Burada A, aktivite konsantrasyonunu (Bq kg⁻¹); m, ürün kütlesini (kg); yaş ve kuru alt indisleri ise kurutma öncesi ve sonrası durumları gösterir. Açık havada kurutmada yüzeye çöken toz da sonucu etkileyebilir. Kaynatmada ise suda çözünen fraksiyonlar pişirme suyuna geçebilir; suyun dökülmesi alımı azaltabilirken yemekle tüketilmesi bu katkıyı korur (Green ve Wilkins, 1996).

5.3. Sütün Peynre Dönüştürülmesi

Peynir üretiminde pıhtılaştırma ve süzme, radyonüklitleri pıhtı ile peynir altı suyu arasında dağıtır. Dağılım, radyonüklidin süt proteinleri ve çözünmüş bileşenlerle etkileşimine bağlıdır.

Süt ve peynir karşılaştırmalarında kullanılan süt miktarı, peynir verimi, nem oranı ve peynir altı suyuna geçen aktivite birlikte değerlendirilmelidir (IAEA, 2010).



Şekil 3. Gıda işleme süreçlerinin doğal radyonüklitlerin aktivite konsantrasyonu ve dağılımı üzerindeki olası etkileri.

5.4. İşleme Etkisinin Değerlendirilmesinde Temel İlkeler

İşleme etkisi, aynı partiye ait ham ve işlenmiş ürünlerin aktivite konsantrasyonları, kütle ve nem değişimleri, kullanılan su ve katkıları ile yan ürünlere geçen aktivite birlikte değerlendirilerek belirlenmelidir. Sonuçlarda yaş veya kuru kütle esaslı belirtilmeli, ürün verimi ve toplam aktivite dengesi dikkate alınmalıdır (Green ve Wilkins, 1996). Tüketiciye yönelik doz değerlendirmesi ise porsiyon miktarı ve tüketim sıklığına dayanmalıdır.

6. GIDA TÜKETİMİNE BAĞLI İÇ DOZ

Gıda tüketimine bağlı iç doz; aktivite konsantrasyonu, tüketim miktarı, yaş ve radyonüklide özgül yutma dozu katsayısına bağlıdır. Aktivite konsantrasyonu tek başına maruziyetin göstergesi değildir.

Az tüketilen yüksek aktiviteli bir ürün, yaygın tüketilen düşük aktiviteli bir temel gıdadan daha az doz katkısı sağlayabilir.

6.1. Yıllık Radyonüklit Alımı ve Etkin Yutma Dozu

Yıllık radyonüklit alımı aşağıdaki eşitlikle hesaplanır:

$$I_i = A_i \times C$$

Burada I_i , i radyonüklidinin yıllık alımını (Bq yıl^{-1}); A_i , gıdadaki aktivite konsantrasyonunu (Bq kg^{-1}); C ise yıllık tüketim miktarını (kg yıl^{-1}) gösterir (IAEA, 2022).

Tüketim verileri mümkün olduğunda bölgeye ve yaş grubuna özgül olmalıdır. Ulusal arz verileri gıda kayıpları ve bireysel farklılıklar nedeniyle gerçek tüketimi tam yansıtmayabilir (IAEA, 2022).

Yıllık etkin yutma dozu aşağıdaki eşitlikle hesaplanır:

$$E_{ing} = \sum_i (A_i \times C \times DCF_i)$$

Burada E_{ing} , yıllık etkin yutma dozunu ($Sv \text{ yıl}^{-1}$); DCF_i , radyonüklit ve yaş grubuna özgü yutma dozu katsayısını ($Sv \text{ Bq}^{-1}$); i ise radyonüklidi gösterir. Katsayılar sindirim sisteminden emilimi, vücut içi dağılımı ve biyolojik uzaklaştırmayı dikkate alır (ICRP, 2012).

^{40}K aktivitesi gıdalarda çoğunlukla yüksek olsa da birim aktivite başına doz katsayısı ^{226}Ra ve ^{232}Th 'ye göre daha düşüktür (ICRP, 2012). Bu nedenle her radyonüklidin doz katkısı ayrı hesaplanmalıdır.

6.2. Gıda Gruplarının Toplam Doza Katkısı

Tahıl ve süt ürünleri yüksek tüketimleri nedeniyle toplam doza önemli katkı sağlayabilirken baharatların düşük tüketimi, yüksek aktiviteye rağmen doz katkısını sınırlayabilir (Bilgici Cengiz ve Çağlar, 2022; Cengiz ve ark., 2026). Bu nedenle gıda izleme çalışmalarında öncelikler; aktivite konsantrasyonu, bölgesel tüketim miktarı, tüketim sıklığı ve hassas yaş grupları birlikte dikkate alınarak belirlenmelidir.

6.3. Yaşa Bağlı İç Doz Farklılıkları

Yaşa bağlı iç doz farklılıkları, tüketim miktarı ve yutma dozu katsayılarının değişmesinden kaynaklanır. Bazı radyonüklitlerin doz katsayıları çocuklarda daha yüksek olsa da nihai doz tüketim miktarına da bağlıdır. Bu nedenle her yaş grubu, kendisine uygun tüketim verileri ve doz katsayılarıyla ayrı değerlendirilmelidir (ICRP, 2012; Bilgici Cengiz ve ark., 2025).

6.4. İç Doz Sonuçlarının Yorumlanması

Hesabın kapsadığı ürünler ve maruziyet yolları açıkça belirtilmeli; sınırlı sayıdaki ürünün dozu bütün beslenmenin toplam iç dozu olarak sunulmamalıdır.

Gıda kaynaklı iç doz, dış ortam gama dozundan farklıdır. Toprak ve yapı malzemeleri için geliştirilen dış doz eşitlikleri gıda tüketimine uygulanmamalıdır (UNSCEAR, 2000; ICRP, 2012).

7. RADYOLOJİK RİSKİN

DEĞERLENDİRİLMESİ VE YORUMLANMASI

Etkin doz doğrudan bir sağlık sonucu değil, farklı maruziyetlerin stokastik etkilerini karşılaştırmak için kullanılan bir radyasyondan korunma büyüklüğüdür (ICRP, 2007).

Yaşam boyu doz ve kanser riski yararlı karşılaştırma göstergeleri olsa da sabit tüketim, yaşam süresi ve düşük doz modeli gibi varsayımlar içerir.

7.1. Yaşam Boyu Etkin Doz ve Kanser Riski

Yıllık etkin yutma dozundan yaşam boyu etkin doz ve yaşam boyu ek kanser riski aşağıdaki eşitliklerle tahmin edilir (Bilgici Cengiz ve Çağlar, 2022):

$$D_{yaşam} = E_{ing} \times T$$

$$ELCR = E_{ing} \times T \times RF$$

Burada $D_{yaşam}$ yaşam boyu etkin dozu (Sv), E_{ing} yıllık etkin yutma dozunu (Sv yıl⁻¹), T maruziyet süresini (yıl), RF ise nominal risk katsayısını (Sv⁻¹) gösterir. RF 'nin kaynağı ve temsil ettiği sağlık sonucu belirtilmelidir (ICRP, 2007). Bu yaklaşım yıllık dozun sabit kaldığını varsayar; daha gerçekçi hesaplamalarda yaşam evreleri ayrı değerlendirilir. ELCR,

bireysel hastalık tahmini değil, varsayımlara dayalı bir karşılaştırma göstergesidir.

7.2. Risk Hesaplarının Sınırlılıkları ve Yaşa Bağlı Değerlendirme

Risk tahminleri; aktivite, tüketim, maruziyet süresi ve doz katsayılarına ilişkin varsayımlardan etkilenir. Doğrusal eşiksiz model radyasyondan korunmada kullanılsa da düşük doz risklerinin epidemiyolojik olarak gösterilmesi güçtür (ICRP, 2007; UNSCEAR, 2012). Çocuk–yetişkin karşılaştırmaları yaşa özgü tüketim ve doz katsayılarına dayanmalı; çocukluk döneminin yıllık dozu tüm yaşam süresine uygulanmamalıdır (ICRP, 2012).

Halk için yıllık 1 mSv etkin doz sınırı, günlük gıdalar için doğrudan bir güvenlik limiti değildir. Bu değerin altında veya üstünde olmak, tek başına kesin güvenlik ya da sağlık etkisi göstergesi olarak yorumlanmamalıdır (ICRP, 2007; IAEA, 2022).

7.3. Doğal Potasyumun Özel Durumu

⁴⁰K, vücuttaki miktarı fizyolojik olarak düzenlenen doğal potasyumun radyoaktif izotopudur (UNSCEAR, 2000). Gıdalardaki yüksek aktivitesi tek başına yüksek risk anlamına gelmez; tüketim miktarı ve doz katsayısıyla birlikte değerlendirilmelidir (ICRP, 2012). Risk değerlendirmesi, maruziyeti karşılaştırmayı, hassas grupları ve izleme önceliklerini belirlemeyi amaçlar.

8. YÖNTEMSEL SINIRLILIKLAR VE ARAŞTIRMA GEREKSİNİMLERİ

Doğal radyonüklitlerin ölçümü ve radyolojik risk hesabı örneklemeden tüketim verisine kadar birbirini izleyen aşamalar içerir. Başlıca belirsizlik kaynakları ve azaltma yaklaşımları Tablo 3'te özetlenmiştir.

Tablo 3. Gıdalarda doğal radyoaktivite ve radyolojik risk değerlendirilmesindeki başlıca belirsizlik kaynakları

Değerlendirme aşaması	Belirsizlik kaynağı	Sonuç üzerindeki olası etkisi	Önerilen yaklaşım
Radyoaktif denge	Numunelerin yetersiz süre veya radon sızdırmazlığı sağlanmamış kaplarda bekletilmesi.	Radon kaybı, ^{214}Pb ve ^{214}Bi üzerinden hesaplanan ^{226}Ra aktivitesinin düşük tahmin edilmesine neden olabilir (Benke ve Kearfott, 2000).	Numuneler radon sızdırmazlığı doğrulanmış kaplarda yeterli süre bekletilmeli ve denge koşulları raporlanmalıdır.
Gama spektrometrisi	Dedektör çözünürlüğü, pik girişimleri, düşük sayım istatistiği ve arka plan	Aktivite konsantrasyonlarının belirsizliği artabilir veya radyonüklitler hatalı tanımlanabilir.	Uygun gama pikleri seçilmeli, arka plan katkısı çıkarılmalı ve mümkünse birden fazla pik değerlendirilmelidir (Bilgici Cengiz, 2020).
Verim kalibrasyonu	Kalibrasyon ve numune arasındaki geometri, yoğunluk ve matris farklılıkları	Foto-pik verimi ve hesaplanan aktivite konsantrasyonu sistematik olarak değişebilir.	Kalibrasyon, numunenin kap geometrisi, dolun yüksekliği ve matris özellikleriyle uyumlu yapılmalıdır (IAEA, 2004).
Tüketim verileri	Ulusal ortalamaların bölgesel, yaşa bağlı ve yüksek tüketici davranışlarını yansıtmaması	Yıllık etkin yutma dozu olduğundan düşük veya yüksek hesaplanabilir.	Bölgeye ve yaş grubuna özgü tüketim verileri kullanılmalı; ortalama ve yüksek tüketici senaryoları ayrı değerlendirilmelidir (IAEA, 2022).
Doz katsayıları	Yaş grubuna veya radyonüklide uygun olmayan katsayıların seçilmesi	İç doz ve yaş grupları arasındaki karşılaştırmalar hatalı olabilir.	Güncel ve yaşa özgü yutma dozu katsayıları kullanılmalı; veri kaynağı açıkça belirtilmelidir (ICRP, 2012).
Yaşam boyu risk	Sabit tüketim, sabit yıllık doz ve tek yaşam süresi varsayımı	Yaşam boyunca değişen tüketim ve doz koşulları göz ardı edilebilir.	Yaşam evrelerine dayalı hesaplama yapılmalı ve risk modelinin varsayımları açıklanmalıdır (ICRP, 2007, 2012).
Ölçüm belirsizliği	Sayım, arka plan, verim kalibrasyonu ve numune kütlesi belirsizlikleri	Aktivite konsantrasyonunun belirsizliği eksik tahmin edilebilir.	Ölçüm belirsizlikleri belirlenmeli ve hata yayılımıyla aktivite konsantrasyonunun birleşik belirsizliği hesaplanmalıdır (IAEA, 2004).

Bu belirsizlikler aktivite konsantrasyonundan yaşam boyu risk hesabına kadar bütün zinciri etkiler. Yöntem, varsayımlar ve belirsizlikler sayısal sonuçlarla birlikte raporlanmalıdır.

8.1. Gelecekteki Araştırma Gereksinimleri

Gelecek çalışmalar; eşleştirilmiş toprak–bitki–yem–hayvansal ürün analizlerine, gıda işlemede aktivite dengesine ve yaşa özgü tüketim verilerine odaklanmalıdır. Standart ölçüm ve raporlama yöntemleriyle yürütülen uzun dönemli izleme, laboratuvarlar arası karşılaştırmalar ve belirsizlikleri içeren doz hesapları önceliklidir. Türkiye’ye özgü ulusal bir radyolojik gıda veri tabanı, sonuçların karşılaştırılabilirliğini güçlendirecektir.

9. SONUÇ

Doğal radyonüklitlerin gıda zincirine transferi; toprak özellikleri, biyoyararlanabilirlik, bitki türü ve hayvan metabolizmasının ortak etkisine bağlıdır. Gıda işleme nihai aktiviteyi uzaklaştırma, yoğunlaşma ve yeniden dağılım yoluyla değiştirebilir. İç doz, aktiviteyle birlikte tüketim miktarı ve yaşa özgü doz katsayılarıyla belirlenirken yaşam boyu risk tahminleri model varsayımları çerçevesinde yorumlanmalıdır.

Eşleştirilmiş örnekleme, işleme öncesi ve sonrası analizler, bölgeye ve yaşa özgü tüketim verileri ile nicel belirsizlik değerlendirmeleri, topraktan sofraya transferin ve tüketici maruziyetinin daha güvenilir biçimde ortaya konulmasını sağlayacaktır.

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THE RELATIONSHIP BETWEEN RADON GAS EMISSION AND EARTHQUAKES

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1. INTRODUCTION

Radioactivity is the process by which unstable atomic nuclei transform into other nuclei without any external input of energy, emitting alpha particles, beta particles or gamma rays. The ionizing radiation released in this process has enough energy to remove electrons from atoms as it interacts with matter. Most of the radiation to which humans are exposed comes from natural sources (UNSCEAR, 2008). Naturally occurring radioactive materials in soil, rocks, water and air, together with cosmic rays from space, continuously expose all living organisms to both external and internal irradiation. For this reason, determining environmental radioactivity levels is important for protecting public health and is also widely used as a tool in the earth sciences.

Among natural radiation sources, radon gas, which forms in the uranium decay chain, holds a special place. Radon is one of the largest contributors to the natural radiation dose received by humans (UNSCEAR, 2008). Because it may respond to stress, crack formation and fluid movement in the Earth's crust, it has also long been studied in earthquake precursor research. This study aims to review research conducted in Türkiye and worldwide on the relationship between radon emission and

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earthquakes, and to set out the potential and limitations of this relationship in light of the current literature.

2. RADON

Radon is a colorless, odorless and tasteless noble gas with atomic number 86. Its melting point is -71 °C and its boiling point is -61.7 °C. Radon is formed by the decay of naturally occurring radioactive elements in rocks, soil and water, and it has three natural isotopes: ^{222}Rn (radon) from the ^{238}U series, ^{220}Rn (thoron) from the ^{232}Th series and ^{219}Rn (actinon) from the ^{235}U series (Sandıkcıoğlu Gümüş, 2024; Woith, 2015). The half-lives of these isotopes are 3.82 days, 55.6 seconds and 3.96 seconds, respectively. Actinon has a very short half-life, and the $^{235}\text{U}/^{238}\text{U}$ ratio in natural uranium is only 0.00725. As a result, its concentration in air is negligibly low (Yeşilbağ, 2008).

The distribution of radon in the environment is closely related to the geological structure of a region. Radon produced in rocks reaches the surface either directly as a gas or dissolved in groundwater, and the average radon concentration in the atmosphere is about 0.1 pCi/L (Özmen, 1995). Radon can be transported over relatively long distances mainly in aqueous media. Because its solubility in water decreases markedly with rising temperature, cold and lukewarm waters generally contain more radon (Özmen, 1995).

In the open atmosphere, radon remains at low levels that depend on weather conditions. In enclosed spaces, however, it can accumulate to high concentrations, which makes it a significant indoor pollutant (Kulalı, 2009). Since people spend about 90% of their time indoors, this poses a serious exposure problem. Most of the radon in buildings comes from the soil and rocks beneath them. Because the pressure of soil gas is usually

higher than the indoor air pressure, radon seeps indoors through cracks and openings in floors and walls (WHO, 2009).

3. THE EARTHQUAKE–RADON RELATIONSHIP

Efforts to forecast earthquakes rely largely on observing precursory phenomena that appear before an earthquake. The main ones include ground deformation, changes in seismic wave velocities, and variations in the geomagnetic field and groundwater level. However, because the physical mechanisms of earthquakes and their precursors are still not fully understood, these methods remain largely empirical. In recent years, anomalies in geogases such as helium, hydrogen and carbon dioxide have also been monitored in earthquake-prone regions; among these gases, radon has stood out because it is easy to detect and has become one of the main focal points of earthquake precursor research (Immè & Morelli, 2012).

Of the three naturally occurring isotopes of radon, ^{222}Rn , ^{220}Rn (thoron) and ^{219}Rn , the ^{222}Rn isotope, which is produced by the decay of ^{226}Ra and has a half-life of 3.8 days, is the most important in these studies because it is long-lived enough to be transported from soil and rocks to the surface (Díaz & Liza, 2025; Oh & Kim, 2015).

The distribution of radon in the environment does not depend on tectonic processes alone. Environmental variables such as the mineralogical composition of rocks and soil, their uranium and radium content, porosity and permeability, groundwater movement, and atmospheric pressure, temperature, precipitation and humidity can all affect radon concentration (Planinić et al., 2004; Woith, 2015; Zmazek et al., 2003).

The idea that changes in the concentration of radon, which reaches the surface from rock and soil pores by diffusion or via carrier fluids (groundwater and gases such as CO₂ and CH₄), may be related to stress and deformation processes in the Earth's crust became a focus of international research after a threefold increase was observed in a deep well before the 1966 Tashkent earthquake (Ulomov & Mavashev, 1967; Woith, 2015). Since then, radon has been investigated as a possible earthquake precursor by monitoring it in soil gas, groundwater, thermal spring water and even indoor air.

3.1. Physical Models of Radon Anomalies

The main physical mechanisms proposed to link radon anomalies with earthquakes are as follows:

(i) The dilatancy–diffusion model is based on the assumption that increasing tectonic stress causes cracks in rock to open or widen, forcing radon accumulated at depth toward the surface (Scholz et al., 1973, as cited in Immè & Morelli, 2012).

(ii) The compression model is based on the assumption that increasing crustal compression before an earthquake squeezes soil gas toward the atmosphere (King, 1978, 1980; Sandıkcıoğlu Gümüş, 2024).

(iii) The stress corrosion model discusses the possible role of slow crack growth driven by stress corrosion in geophysical processes. In the radon–earthquake literature, this approach has been used to establish a possible link between slow crack growth and radon transport, particularly in water-saturated rocks (Anderson & Grew, 1977).

(iv) Mathematical transport models and empirical criteria: Immè and Morelli (2012) also formulated radon transport using mathematical diffusion equations such as the “plane sheet model” and the “infinite source model,” and compiled various empirical

formulas from the literature for estimating the magnitude–time–distance relationship (Dobrovolsky et al., 1979; Martinelli, 1992; Rikitake, 1987). For example, according to the formula of Dobrovolsky et al. (1979), the maximum distance at which precursory signs of an earthquake can be observed is calculated as $d = 10^{0.43M}$ km; this formula has been used as an earthquake screening criterion in many subsequent studies (İnan et al., 2012; Kim et al., 2018).

4. PREVIOUS STUDIES

Woith (2015) carried out a comprehensive review of the literature on the radon–earthquake relationship. The study examined 147 time series from 87 studies published between 1967 and 2014 and showed that radon anomalies were reported at a higher rate in short observation series than in longer ones. About 75% of the time series were shorter than three years and only 19% were longer than five years, and in roughly half of the series only a single radon anomaly was reported. Woith also stressed that non-tectonic processes can produce radon variations that closely resemble tectonic anomalies. The study therefore shows that not all anomalies reported in the literature can be attributed to earthquake preparation processes and that long-term monitoring and the separation of environmental and anthropogenic effects are critical. Throughout this chapter, earthquake magnitudes are given as reported in the original studies: M_w denotes moment magnitude, M_L local magnitude, and M is used where the original study did not specify the scale.

4.1. Studies in Türkiye

With its active fault systems and diverse geological and hydrogeological settings, Türkiye has been an important study area for investigating the radon–earthquake relationship. Various studies on soil-gas and groundwater radon have been conducted,

particularly along the North and East Anatolian fault systems and in the active fault zones of Western Anatolia (Erees et al., 2007; Friedmann et al., 1988; İnceöz et al., 2006). While these studies indicate that radon distribution may be related to fault zones, they also show that radon variations cannot be explained by tectonic processes alone.

İnan et al. (2012) carried out continuous soil-gas radon measurements for more than three years at three stations in the Eastern Mediterranean Region of southern Türkiye and showed that radon values exhibit marked seasonal variations. Radon values were found to be higher between May and October and lower between November and April. The researchers emphasized that evaluating precipitation data together with radon data makes it easier to interpret anomalies and that, in long-term measurements, it is important to establish the characteristic baseline behavior of each station. The study stated that seasonal and meteorological variability should be determined before radon anomalies are linked to earthquakes.

Saç et al. (2011) monitored soil-gas radon at four stations along the Tuzla Fault in western Türkiye and used an online continuous measurement system at one of them. Measurements obtained with LR-115 nuclear track detectors were compared with regional seismic data, and the researchers reported a linear relationship between radon emission and seismic activity in the study area. This result is important in showing that continuous radon monitoring in a particular active fault zone can be evaluated together with seismic activity; however, it was noted that caution is needed in generalizing the findings directly to other regions because they pertain to a specific study area.

In a study conducted by İnceöz et al. (2006) on the North and East Anatolian active fault systems, the distribution of soil-gas radon was examined using CR-39 solid-state nuclear track

detectors. The study reported that radon concentrations over the fault zones were higher than in the surrounding areas and that radon values decreased markedly with distance from the fault line. This finding indicates that active fault zones may behave differently in terms of radon transport and that the position of the measurement point relative to the fault is important for interpreting the results.

Baykara and Doğru (2006) measured radon and radium in various water samples and uranium in soil samples along the North and East Anatolian fault systems. The study determined radon and radium levels and radon exhalation rates in the water samples and also examined uranium concentrations in the soils. Rather than being a direct test of earthquake precursors, this study contributes to characterizing the geological and hydrogeological distribution of radon in active fault systems.

Yalım et al. (2012) monitored groundwater radon concentrations at two-week intervals between August 2009 and September 2010 in four wells on the Akşehir–Simav Fault System in Afyonkarahisar and investigated the relationship between radon anomalies and earthquake magnitude. Anomalous decreases in radon concentration were observed in the wells before earthquakes of magnitude M_w 2.6–3.9. The R^2 values for the relationship between radon minima and earthquake magnitude in the four wells were 0.79, 0.93, 0.98 and 0.90, respectively. The researchers interpreted these results as indicating that groundwater radon at measurement sites with suitable hydrogeological conditions may be sensitive to strain changes in the crust. Nevertheless, because the study was based on four wells and an observation period of about one year, and because the earthquakes examined were relatively small, it was noted that longer-term observations are needed to establish whether the relationship can be generalized to larger earthquakes and different hydrogeological settings. In addition, given that the high R^2

values rely on a limited number of earthquakes for each well, the statistical reliability of these coefficients needs to be tested with larger data sets.

Groundwater radon monitoring can reveal anomalies that differ from those found in soil-gas measurements. Sandıkcıoğlu Gümüş (2024) measured radon in six deep water wells on the Akşehir–Simav Fault System for 24 months and reported that before some earthquakes the radon concentration dropped below the background level and then returned to the background level or rose above it. This behavior, described in the literature as a “V-shaped” anomaly, has been associated with earlier studies in Taiwan (Kuo, 2014); however, it was noted that this pattern did not appear clearly in Sandıkcıoğlu Gümüş’s own measurements. The study obtained high correlation coefficients ($R^2 = 0.82\text{--}0.97$) between radon minima and earthquake magnitude in all six wells. Nevertheless, the fact that radon anomalies were not observed in every well before every earthquake, and that some anomalies were not followed by any earthquake, shows that the observed relationship is not sufficient on its own for earthquake prediction. Thus, while the study shows that changes in groundwater radon may be related to earthquake processes, it also highlights the importance of long-term monitoring and of considering other hydrogeological parameters when evaluating this relationship.

Keskin and Külahcı (2023) examined soil ^{222}Rn variations before the M_w 4.2 Sivas–Suşehri earthquake, which occurred on the North Anatolian Fault Zone in 2007, together with earthquake activity and atmospheric total electron content (TEC). The study investigated the relationship between radon concentration and microseismic activity and also included meteorological variables in the analysis, such as soil temperature at depths of 5, 10, 20 and 50 cm, vapor pressure, and wet-bulb and dry-bulb temperatures. The researchers reported that relative radon concentration and TEC increased together as the main shock approached and that

radon was correlated with some of the meteorological variables. In addition, they aimed to forecast future radon concentrations using an ARIMA(4,1,3) model. The study is noteworthy for evaluating the radon–earthquake relationship not only through the radon time series but also by combining seismic, atmospheric and statistical time-series data. Nevertheless, the validity of the relationships it reports for other earthquakes and regions needs further investigation.

Taken together, the studies conducted in Türkiye show that the radon–earthquake relationship cannot be explained by a single type of observation. Soil-gas measurements can provide information on the characteristics of fault zones, long-term continuous monitoring highlights the importance of seasonal and meteorological variations, and groundwater studies require radon behavior to be evaluated in light of hydrogeological conditions. The Turkish literature therefore shows that, for radon to be used in earthquake research, the characteristics of the measurement setting, long-term baseline variability and environmental effects must all be considered together.

4.2. Studies in Other Countries

Liu et al. (2025) is one of the recent studies that quantitatively examine the physical mechanisms of earthquake-related changes in groundwater radon. The researchers jointly analyzed radon concentration, water discharge and water temperature data from the Banglazhang #1 hot spring in the Yunnan region of China after the 1996 Lijiang (M_w 7.0) and 2004 Sumatra (M_w 9.1) earthquakes. The analysis showed that meteorological factors were not the main cause of the changes in radon concentration. The main mechanism of the radon change after the Lijiang earthquake was found to be a change in the mixing ratio of waters from different depths, whereas after the Sumatra earthquake, radon release through the movement of rock

particles may also have played a role in addition to the change in mixing ratio. The water–rock interaction surface area after the Sumatra earthquake was calculated to have increased from approximately $7 \times 10^4 \text{ m}^2$ to $1.85 \times 10^5 \text{ m}^2$. The main conclusion of the study is that the joint evaluation of discharge, water temperature and radon data can provide a stronger basis for explaining earthquake-induced hydrological changes. The researchers therefore stress that multiparameter monitoring is necessary for earthquake-related radon signals to be evaluated accurately and in a timely manner. It should be borne in mind, however, that the study examined post-earthquake rather than pre-earthquake radon changes; it therefore contributes to understanding the mechanisms of earthquake-related radon changes rather than serving as a direct test of precursors.

Oh and Kim (2015) took a different approach to the radon–earthquake relationship by monitoring ^{222}Rn and ^{220}Rn (thoron) together in a limestone cave in South Korea. They noted that, because ^{220}Rn has a half-life of only 55.6 seconds, it is less affected by environmental factors and may allow short-range radon variations to be tracked. The researchers monitored ^{220}Rn and ^{222}Rn concentrations in the cave air for one year and observed unusually high ^{220}Rn peaks in February, before the 2011 Tohoku-Oki earthquake (M_w 9.0). By contrast, high ^{222}Rn values were found both in February and during the summer. The researchers suggested that the ^{222}Rn and ^{220}Rn anomalies observed in February may have been precursory signals related to the Tohoku-Oki earthquake. The study shows that monitoring the ^{220}Rn – ^{222}Rn isotope pair together, rather than ^{222}Rn alone, can provide additional information for distinguishing changes caused by environmental effects and for evaluating radon anomalies with respect to earthquake processes.

To examine the radon–earthquake relationship in an indoor setting, Kim et al. (2018) carried out continuous indoor

radon measurements between February 2016 and January 2017 in a building of Dongguk University in Gyeongju, South Korea. Measurements were made at 30-minute intervals with a RAD7 detector in the Energy Engineering Hall, on the ground floor of the building, following the EPA measurement protocol. The study focused in particular on the M_L 5.1 and M_L 5.8 earthquakes of 12 September 2016 and their aftershocks. The researchers reported that, when the interval between successive earthquakes was long enough, indoor radon concentrations showed similar patterns of sudden increase and decrease around the earthquakes. In some events, this pattern consisted of a sudden rise in radon concentration 1–4 days before the earthquake, followed by a gradual decline before the earthquake and a sudden drop on the day of the earthquake. The study noted, for example, that the pattern was more pronounced when successive earthquakes were about three days apart.

Reddy and Nagabhushanam (2011) monitored groundwater electrical conductivity (EC), groundwater level and soil-gas radon hourly for about a year in the Koyna–Warna region of India, which is known for reservoir-triggered seismicity. They examined the responses to the M 4.7 (14 November 2009) and M 5.1 (12 December 2009) earthquakes and found that EC changes varied from well to well and from one earthquake to the other. For example, in the Govare well a change began about 40 hours before the M 5.1 earthquake, whereas no clear precursory signal was detected in the Koyna well only 4 km away. Soil-gas radon, by contrast, behaved similarly for both earthquakes. It began to rise about 20 days in advance and then fell during the earthquakes by about 30% for the M 4.7 event and about 50% for the M 5.1 event. The researchers attributed the radon anomalies to the opening and closing of microcracks. The fact that wells so close together responded differently indicates that the anomalies may be sensitive to local geological and hydrogeological conditions.

In long-term monitoring in Amritsar, India, Ramola et al. (1990) reported that soil-gas radon was strongly correlated with temperature, pressure, wind, precipitation and humidity. Nevertheless, sudden radon increases were also observed during periods of normal meteorological conditions. Some of these were followed by earthquakes of magnitude M 3.8–5.7 that occurred 150–400 km away in 1986–1987. The authors emphasized that there were also anomalies that could not be linked to earthquakes. They stated that objective criteria need to be developed to remove meteorological effects and instrumental artifacts.

Nikolopoulos et al. (2024) reviewed some 40 years of research on electromagnetic and radon precursors in a comprehensive review. According to the authors, radon is one of the longest-used precursors in earthquake research. Because it is inert and can be transported over long distances, it may also offer an advantage for monitoring distant earthquakes. The review notes that anomalies are still mostly identified visually or with the $\pm 2\sigma$ criterion. In recent years, however, methods such as natural time analysis, DFA, fractal dimension, the Hurst exponent and entropy-based approaches have become more common. The authors stress that, because every earthquake is unique, it is difficult to build a universal model. They also point out that the retrospective identification of most precursors after the earthquake has occurred is a major limitation.

Planinić, Radolić and Vuković (2004) monitored soil-gas and well radon with LR-115 detectors at five sites in Croatia for four years (1998–2002), together with barometric pressure, precipitation and temperature. Meteorological effects were modeled by multiple regression ($R^2 = 0.46$; $F = 6.17 > 3.71$, $p < .05$). The variations remaining after these effects were removed were evaluated as anomalies. Of the 26 earthquakes of magnitude M 2.5–4.5 recorded during this period, 58% were reported to be recognizable from radon anomalies, as were all earthquakes with

$M \geq 3$ located within 200 km. A key strength of the study is that meteorological effects were removed statistically before anomalies were defined. However, because the false alarm rate was not reported, the 58% recognition rate alone does not reflect the method's predictive success.

Ghosh, Deb and Sengupta (2009) evaluated studies worldwide on the use of radon in earthquake research in a comprehensive review. Along with radon anomalies observed in soil gas, groundwater and spring water, the review covers various empirical and theoretical models developed to explain the relationships between radon anomalies and earthquake magnitude, epicentral distance and precursor time. The review also includes the Turkish studies of Friedmann et al. (1988) and İnceöz et al. (2006). In this respect, it is a useful source for assessing the development of the literature, as it brings together different observational findings and modeling approaches to the radon–earthquake relationship. For a case study on the use of radon as an earthquake precursor in the northwestern Himalayan region of India, see also Walia et al. (2005).

Zmazek et al. (2003) used various regression methods to predict soil-gas radon measured at three stations in the Krško Basin of Slovenia from barometric pressure, air and soil temperature and precipitation. Of these methods, model trees performed best. During seismically quiet periods, radon could be predicted from environmental parameters with a correlation above 0.8. About 1–7 days before earthquakes of M_L 0.8–3.3, however, the agreement between measured and predicted values deteriorated markedly. In this approach, an anomaly is defined not as an increase in radon but as a drop in the model's predictive performance. Indeed, radon increased before some earthquakes and decreased before others. The authors emphasized that longer observations are needed to explain this differing behavior and its relationship with magnitude.

Alam et al. (2020) examined the effect of the 12 May 2008 Wenchuan earthquake (M_w 7.9) on groundwater radon at nine stations. Hurst analysis showed that the series were persistent ($0.5 \leq H \leq 1$) rather than random. Anomalies were identified with the $\pm 2\sigma$ criterion on the residual signal obtained by removing the seven-day moving average. Anomalies were observed at seven stations near the earthquake as well as at two stations about 1,550 km from the epicenter. At these distant stations, the anomalies appeared 11–15 days before the earthquake. The study also reported a decrease in GPS-TEC data over the epicenter and interpreted this as possible support for the Lithosphere–Atmosphere–Ionosphere Coupling (LAIC) model. The authors stated that their findings suggest radon may be a promising tool in earthquake prediction research.

5. CONCLUSION

The idea that radon could serve as an earthquake precursor became a subject of international research after a marked increase in radon was observed in a deep well before the 1966 Tashkent earthquake, and it has since given rise to a body of literature spanning some sixty years. During this period, radon has been monitored in a variety of settings, including soil gas, groundwater, thermal springs and indoor air, and researchers have investigated whether changes in radon concentration are related to stress, deformation, crack formation and fluid movement in the Earth's crust. Existing studies show that, under certain geological, hydrogeological and tectonic conditions, radon anomalies can be linked temporally or spatially to some earthquakes. However, it has not yet been demonstrated that this relationship is strong enough to support a generalizable and reliable early warning system.

One of the main reasons for this uncertainty is that radon concentration is influenced by many environmental and physical factors. In addition to meteorological conditions, geological structure, permeability, groundwater movement and the characteristics of the measurement setting can also cause variations in radon concentration. In some cases, these variations can resemble anomalies thought to be earthquake-related. Therefore, attributing an increase or decrease in radon concentration to an earthquake on that basis alone is not sufficient. Considerable uncertainty arises particularly from short-term measurements and from retrospectively matching a single anomaly with a specific earthquake that occurred later. Long-term measurements, by contrast, provide a stronger basis for establishing the normal radon behavior of a region and for evaluating unusual variations more reliably.

Some studies in the literature report high correlation coefficients or high recognition rates. However, many of these results are based on a small number of stations, short observation periods and a limited number of relatively small earthquakes. Consequently, although these findings are meaningful for the specific measurement sites concerned, they cannot be directly generalized to other regions or to larger earthquakes.

Taken together, the studies in the literature indicate that more reliable results can be obtained when radon data are considered together with the characteristics of the measurement setting and other independent variables. In studies of groundwater and thermal springs in particular, evaluating radon variations together with water temperature, discharge, water level and other hydrogeochemical parameters helps clarify the likely source of the observed anomalies. In this way, rather than being attributed directly to a tectonic process, a change in radon concentration can be evaluated alongside other physical and hydrogeological changes occurring during the same period.

In recent years, statistical methods have also become increasingly important in radon studies. The use of methods such as multiple regression, model trees, time-series models such as ARIMA, the Hurst exponent and fractal analyses helps separate meteorological and other environmental effects and allows observed variations to be evaluated more objectively. In addition, multiparameter approaches, such as the joint monitoring of the radon–thoron isotope pair or the evaluation of radon together with hydrological parameters, are increasingly common in the literature.

In conclusion, some sixty years of research have produced a large body of observational data pointing to a potential relationship between radon and seismic processes. Current findings suggest that, under suitable conditions and when evaluated together with other independent indicators, radon can be regarded as a geochemical indicator that may contribute to monitoring changes in the Earth’s crust, particularly in fluid systems. Future studies should aim to explain the physical mechanisms that give rise to anomalies and to distinguish these changes from environmental effects. They should also investigate whether the findings can be reproduced in different regions and geological settings.

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**FİZİK ALANINDA GÜNCEL KONULAR VE
AKADEMİK DEĞERLENDİRMELER**

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