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Simulation tools for improvement of the fission track analysis method for nuclear forensics

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Abstract

This research introduces an innovative simulation software for nuclear forensics modeling. The software simulates fission tracks based on physical and radiation parameters, resembling light microscope images. These simulated tracks serve as foundational data for AI-driven decoder software and image analysis. The simulator also functions as an operator trainer and performance assessment tool. By connecting theory and application, it enhances nuclear forensics investigations, contributing to nuclear security and nonproliferation efforts. This versatile tool holds promise for deeper insights and robust methodologies in nuclear forensics, emphasizing the necessity of comprehensive validation using real-world data as a crucial focus for future research.

Keywords Nuclear forensics · Fission track analysis · Enriched uranium · Homeland security · Safeguards investigations

Introduction

Nuclear forensics and safeguards investigations rely heavily on the physical, chemical, and isotopic analysis of individual particles containing fissile isotopes like ^{235}U. These particles of interest (POIs) are often present in mixtures with numerous other particles, such as soil or dust. To conduct examinations using advanced methods like mass spectrometry measurements, the POIs must first be identified and separated. One of the most used techniques for identifying POIs is fission track analysis (FTA), analyzing the fission tracks of fissile particles [1]. Other known techniques are neutron activation imaging [2], nuclear spectroscopy, and time of flight secondary ion mass spectrometry (TOF–SIMS) [3]. This paper presents a new software tool that we developed for the global nuclear forensics community to improve fission track image detection and identification. The software is an interactive application that utilizes a versatile database based on Monte Carlo simulations. It can be used for both nuclear forensics research and training operator personnel.

Variants of the FTA methodology have been used for decades in many fields, such as radiometry, nuclear forensics, safeguards investigations, geology, and cosmology [4]. A fission-track (FT) is a microscopic-scale radiation-damaged site, which can be induced on a solid-state nuclear track detector – SSNTD (e.g., polycarbonate sheet, mica, etc.) [5, 6] by a radiant array of nuclear fissions of a single particle under thermal neutron flux. In nuclear forensics, the tracks are developed and made visible by chemical etching of the SSNTD. Since an FT cluster arises from a single particle, it exhibits a stochastically round shape, which defines both the presence and the location of a POI. When detected with transparent or translucent SSNTDs, FTs have optical characteristics that enable their visualization and identification using transmitted or reflected light microscopy [4], as shown in Fig. 1.

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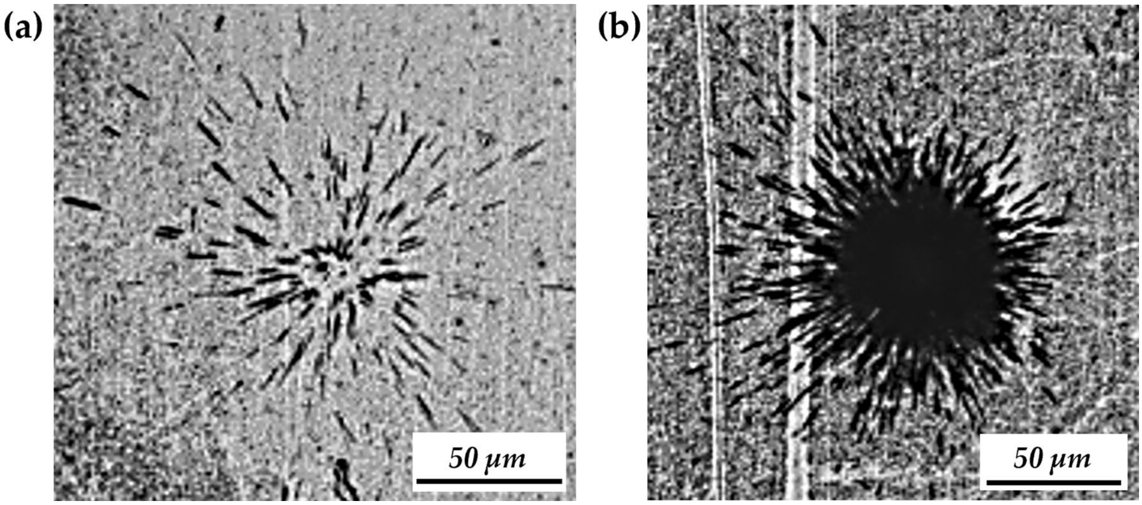


Fig. 1 Fission tracks cluster of different particles for the same SSNTD detector and radiation conditions. **a** Poor cluster and **b** Rich cluster

Two primary approaches are commonly employed for the analysis of fissile isotope ratios: bulk analysis [3], often referred to as "mini bulk," and particle analysis, known as "micro-bulk" [7] We define bulk analysis as the process in which the entire sample is dissolved, and average isotope ratios are determined to encounter the average enrichment using techniques such as Thermal Ionization Mass Spectrometry (TIMS) or Inductively Coupled Plasma Mass Spectrometry (ICPMS). This method offers a macroscopic view of the sample, providing insights into the overall composition. Conversely, we define particle analysis as the calculation of individual fissile isotope ratios of the same particle for detailed insights into its origin. By examining individual particles within the sample, particle analysis allows researchers to uncover fine-scale variations and anomalies in isotope ratios, shedding light on specific source materials or processes. It is a powerful tool for gaining a more granular understanding of the sample's composition and its historical or geological context.

Typically, researchers perform a bulk analysis initially to estimate the quantity of fissile material present, as a precursor to subsequent micro-bulk analysis. The Fission Track Analysis (FTA) method is a representative approach for particle analysis, focusing on identifying the presence of fissile material and estimating its location. However, it's important to note that FTA alone cannot be used to accurately gauge the enrichment of the material. For a comprehensive assessment of isotopic compositions, advanced methods such as ICPMS are required.

The method usually used when performing FTA particle analysis is dissolving the material containing the test particles in the LEXAN® foil, called a "catcher", and inserting it between two SSNTD detectors in the "sandwich formation”.

To clarify the distinction between bulk and particle analysis, a "cutting" procedure is frequently employed on the "catcher". This procedure entails the isolation and dissolution of a single particle (POI), as identified by Fission Track Analysis (FTA), for subsequent analysis via mass spectrometry techniques.

In the field of nuclear forensics, our research group is currently developing innovative techniques designed to enhance the reliability and precision of our analyses. These novel approaches are designed to provide more accurate and robust answers to nuclear forensics questions. Currently, only trained operators can analyze microscope images of FTA data. Since this analysis depends on the operator's judgment and skills, it is obvious that different operators will produce slightly distinct results. A new operator's training period is lengthy, and it requires using numerous examples from previously measured data and some that we can only predict. The software has proven invaluable for assessing the proficiency of new operators. Our software, FTA Trainer V2.0, utilizes Monte-Carlo simulations results, specifically GEANT4, to model fission tracks. It takes into account critical parameters, such as thermal neutron flux, fission cross-section, radiation time, particle size, and enrichment, which collectively determine nuclear fission track characteristics. This user-friendly application calculates fission tracks on a simulated LEXAN® detector and their respective projections, allowing customization of physical parameters and configurations by the user. Developed with MATLAB App Designer infrastructure [8], FTA Trainer V2.0 is compatible with any Windows 10 computer, requiring no specialized setup. Computer requirements are shown in Appendix A.

An example of a software tool developed for modeling fission tracks for geological purposes is discussed by Richard A. Ketcham et al. [9]. It's worth noting that this particular software exclusively focuses on modeling spontaneous fissions and does not encompass induced fissions, which is a key distinction from our software tool.

Theory

Theoretically, the track's length on the SSNTD depends on the particle's kinetic energy, particle type, electric charge, and mass. Fission products are generated when an atomic nucleus splits into different nuclei with varying atomic masses. In our case, focusing on ^{235}U, ^{233}U and ^{239}Pu, the shape of this distribution can change depending on the energies of the free neutrons that initiate the reaction, at thermal energies, where $E \cong 0.025$eV, the fission product mass distribution exhibits two distinct peaks, centered at approximately 90 AMU for light fragments and approximately 140 AMU for heavy fragments. In other fissile isotopes, whether the fission induced by neutrons or spontaneously, the peaks centered differently. In addition to differences in peak placement, each fissile isotope may also exhibit variations in the energy yield.

Due to the unique fission product yields associated with each fissile isotope, the placement of peaks in the mass distribution is a crucial distinguishing factor. However, the FTA method currently utilized for identifying fissile isotopes in a sample does not differentiate between different isotopes or consider the influence of their respective peak distributions in the analysis process. Figure 2 shows the energy distribution of fission products produced by the thermal fission of ^{235}U. The calculated mean energy of the light fragment is approximately 99 MeV, while the mean energy of the heavy fragment is approximately 68 MeV [10]. In theory, the analysis of track lengths on SSNTDs can be employed to distinguish between different isotopes.

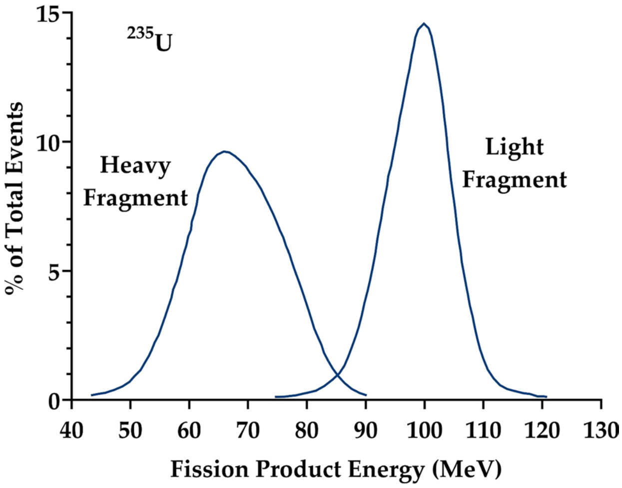


Fig. 2 Energy distribution of fission products produced by the thermal fission of.^{235}U [10]

Experimental

In our comprehensive investigation of nuclear fission behavior, we seamlessly integrated Monte Carlo simulations using GEANT4, to examine the behavior of fissile isotopes within single particles, alongside the development of the Fission Track Analysis Trainer Application (FTAT). The Monte Carlo simulations served as the foundation for our understanding of fissile materials at the particle level, allowing us to explore various configurable parameters. Building on these insights, we crafted FTAT using MATLAB App Designer, which generates synthetic fission track models exported as an image data based on the simulation data. Furthermore, to validate the real-world applicability of FTAT, we conducted laboratory experiments with IAEA-314 Stream Sediment reference materials, focusing on the impact of different etching times on track width. The results of these experiments were integrated into FTAT, enhancing the software's adaptability and alignment with practical conditions.

Monte-Carlo simulations

To deeply study the behavior of nuclear fissions, a simulation was performed examining variations of single particles (grains) containing fissile isotopes (e.g., ^{235}U & ^{233}U) using Monte Carlo simulation.

The simulation platform used in this work is GEANT4 [11] version 10.6 with the "QGSP_BERT_HP" physics library and utilizing the "G4NEUTRONHP_PRODUCE_FISSION_FRAGMENTS" environment variable.

Each simulation maintained a consistent primary geometry. This geometry consisted of a world volume shaped as a cubic region with dimensions of 10 cm^3, filled with air. In the center of this world volume, a layer of LEXAN® plastic measuring 3 cm in width and height, and 1615 µm in thickness, was placed.

In the center of the LEXAN® layer, we placed the fissile particle which is surrounded by a spherical isotropic neutron source resembles the process of irradiating real sample in a nuclear reactor. Therefore, the neutron flux was homogeneous, without preferred orientation and directed inward the particle from all directions (4π). The energy of the neutrons was monoenergetic with 0.025 eV (thermal neutrons). By placing the fissile particle in the center of a thick LEXAN® layer, it created a "sandwich" configuration resembling the arrangement of two SSNTD detectors.

Notably, our simulation offered the flexibility to adjust various configurable parameters, including:

- Total Particle Diameter in [µm].
- Particle Geometry (Spherical or Cylindrical).
- Thermal Neutron Flux in [#/cm^{2}•s] and Irradiation Time of neutrons in [s] both derived from the total neutron events demanded.
- Isotope Enrichment [%].
- Different Fissile Isotopes.
- Natural Soil content in the particle [g].

This setup allowed us to explore a wide range of scenarios, providing valuable insights into the behavior of nuclear fission under different conditions.

Figure 3 presents a visualization of the irradiation simulation process featuring a spherical ^{235}U particle with a 1 µm diameter, positioned in the LEXAN® layer which effectively functions as two SSNTD detectors combined together. The number of neutrons used in this specific simulation was 2000. For the visualization, we have utilized the Open Graphics Library (OGL) which is a powerful rendering library employed by GEANT4 to display 3D graphics. In addition, the simulation was integrated with QT framework, allowing us to view the simulation results and interact with the geometry through a QT window.

The simulations initiated the fission process within the irradiated particle, resulting with a production of fission products. These fission products subsequently interacted with the LEXAN® layer. The trajectories of each fission product were monitored and recorded in a database log.

Numerous databases were generated through simulations employing various parameters, capturing the precise trajectories of fission products resulting from the nuclear fission process. The primary objective of these Monte Carlo simulations was to produce physical data, which is then extracted into '.csv' files for subsequent analysis using our dedicated application software—Fission Track Analysis (FTA) Trainer.

Fission track analysis trainer application

To gain a deeper understanding of the impact of the fissile particle configuration on the fission tracks generated in SSNTD detectors, an analysis tool is required. Therefore, we developed a dedicated application—the FTA Trainer. This application can generate synthetic models of fission tracks, using the fission products trajectory data base, which was created by GEANT4 simulations, and creates a bank of image data, resembling light microscope images.

The application software was developed using MATLAB App Designer [8]. The App is exported as a Windows executable file and can run on almost any Windows-based computer without any specific environment or infrastructure. Calculations can be made based on accurate real data. By entering the diameter of the Fissile Material, Thermal neutron flux, Fission cross-section (Selectable between the dominant fissionable isotopes: ^{235}U, ^{233}U, and ^{239}Pu), neutron irradiation time, and Fissile material mass, it is possible to calculate the number of tracks that are produced in a simulated Cluster.

Projection analysis of the fission product paths is added to the software for studying deeply the behavior of nuclear fission products when making an interaction with the SSNTD Detectors, which are placed in a "sandwich configuration" containing the particle [5], see Appendix B. The simulation shows the spatial behavior of the fission products and the plane projection. In addition, it emphasizes the different appearance of the clusters depending on the distance between the fissile material and the projected plane in the

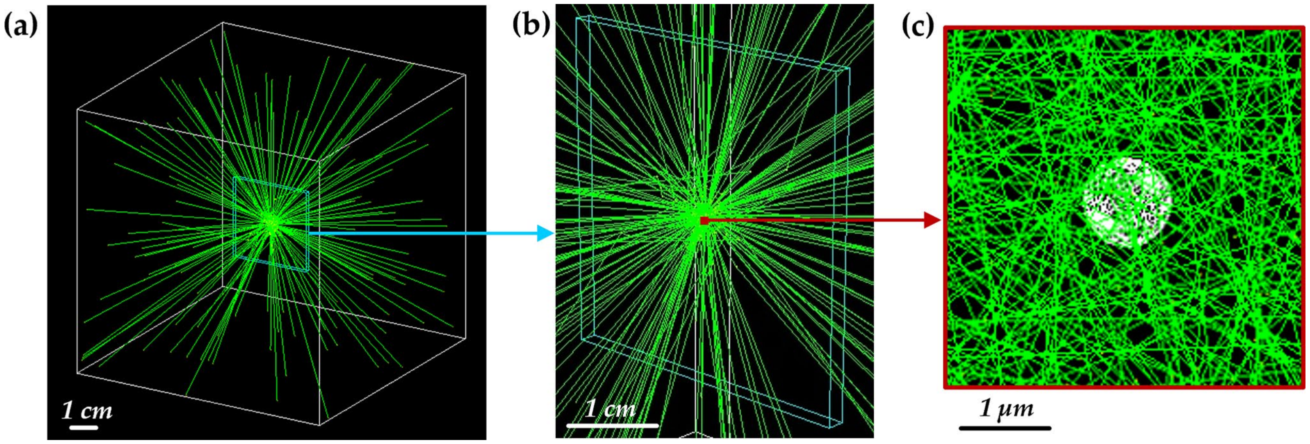


Fig. 3 **a** Visualization of the world volume shaped as a cubic region and the thermal neutron flux (green path lines) with energy of 0.025 eV interacting with a 1 µm particle of natural uranium placed in **b** LEXAN® layer which acts as two combined SSNTD detectors. **c** The spherical shape of the irradiated particle

LEXAN®. A 3D analysis is available to map the exact sites of fissions in the simulated particle (Appendix B).

Single cluster is a super-position of fission track that share the same starting point which is the irradiated particle. Comparison between real and simulated single clusters is shown in Fig. 4. The simulated single clusters (Red centered) and the original single clusters look similar.

In our research, our focus has been on enhancing the FTA Trainer, elevating it into a versatile tool capable of replicating a diverse range of fission track cluster image data, resembling microscope images.

During the replication of fission track models, our application undergoes a series of intricate steps. It begins with the selection of a clear background image to serve as the canvas. The core functionality of the application relies on a meticulously curated database containing trajectory data from GEANT4 simulations, detailing the paths of fission products. These virtual trajectories are then fused with mathematical principles and physics equations, which we have embedded directly into the application. The precise physics data further informs the modeling process. The application's algorithm uses trigonometry and other mathematical methods to transform this data into versatile Fission Track models that cater to the user's specific demands. We implemented mechanisms to ensure the accuracy and realism of the generated Fission Track models and compared the output images with expected results and experimental data to validate the accuracy of the simulations as shown in Fig. 4. The outcome represents a bridge between cutting-edge simulation data and the production of high-fidelity microscope-like images. Refer to Appendix C for a comprehensive overview of the main steps in the development process, the complete list of software capabilities, and a code example illustrating the tracing of fission product paths.

To demonstrate the relation of the fission products energy to the track's lengths, a histogram analysis was added to the software to count the number of tracks per length. When thermal neutrons are used in the simulations ($E \cong 0.025\text{eV}$), the fission products are generated with two typical kinetic energies. Therefore, if a histogram analysis is performed, most of the track's lengths will be contained in a bimodal Gaussian distribution, whose peaks will be determined by the typical energies, as shown in Fig. 5.

The analysis of track lengths, using the developed FTA Trainer software, enables the investigation of their potential to distinguish between different isotopes, in alignment with existing theoretical principles.

Additionally, FTA Trainer also functions as an operator trainer and performance assessment tool for grading the judgment and abilities of operators performing cutting operations at Points of Interest (POIs).

The data generated through these simulations serves as supplementary information for researchers. Its primary utility lies in research purposes, offering insights into a wide range of conditions and providing a valuable resource for training operators. Looking ahead, our research group is working on leveraging this data to develop AI-driven decoder software and image analysis tools, aiming to automate the identification of clusters without human intervention and replace the manual methods. This automation ultimately seeks to reduce the likelihood of human errors that can occur during manual procedures. However, it's important to note that these topics are beyond the scope of this paper.

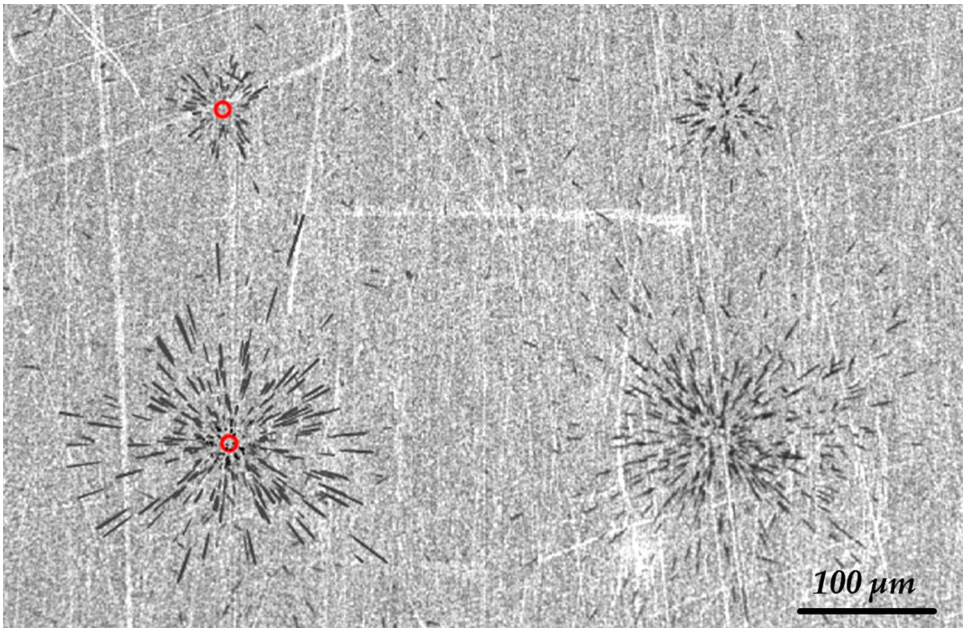


Fig. 4 Comparison between real and simulated (Red centered) Single Clusters

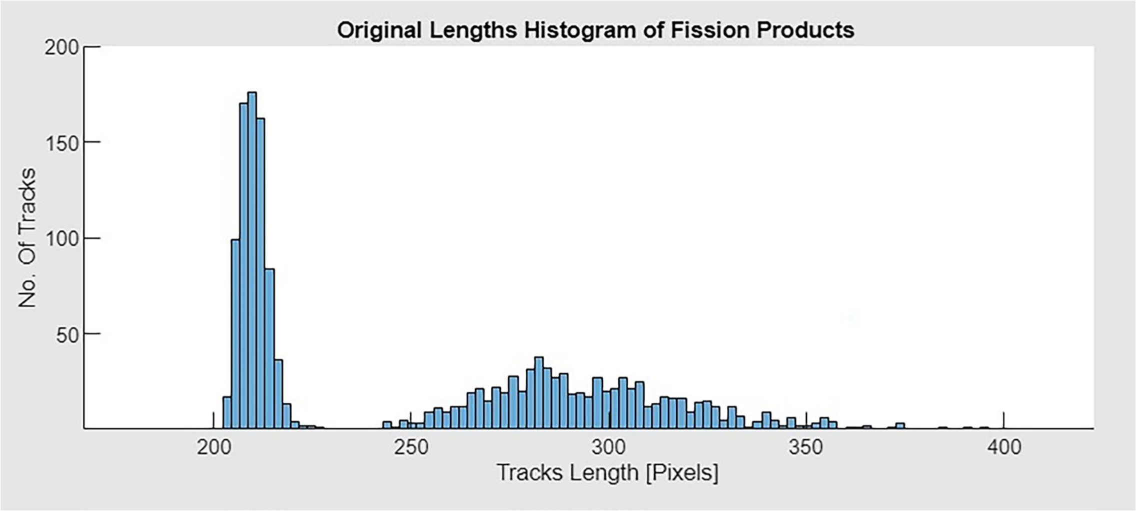


Fig. 5 Fission Tracks Lengths Histogram Analysis made by FTA Trainer for 1 µm ^{235}U particle with ~ 1000 fissions

Fission track width data analysis

The FTA method utilizes a manual, time-sensitive chemical etching process to reveal FT clusters from particles of interest (POIs). In our study, using LEXAN® SSNTD, we examined the impact of different etching times on the actual track width and integrated these findings into the FTA Trainer software application. This enhancement enhances the software's flexibility, making it more closely aligned with real-world conditions.

The materials used for the experiment were IAEA-314 Stream Sediment reference materials [12]. In the experiment, two LEXAN® SSNTDs were used with a natural uranium foil. The detectors and the natural uranium foil were held in a specially designed holder, with the uranium sandwiched between the two detectors. Am-Be was used as the source of neutrons. The source intensity was 10^6 n/sec, and the exposure time was one week. The upper detector was etched in a 6N NaOH solution, at 70°C, for 13 min (The bottom detector was not etched). The etching served to “develop” the FT clusters, making them visible under light microscopy [2, 5]. After the etching process, the etched detector was viewed with a Nikon Eclipse Ni microscope, see the set up used in Appendix D. Using this microscope and camera, a digital image of the detector was generated. Single track width was measured using the Gwyddion-2.60 open-source software package [13]. At least 30 Tracks were measured and averaged. The process was repeated ten times to determine the dependence of the track width on the etching time. After 130 min, the optical quality of the detector decreased due to over-etching of the surface.

Usually, a thermal neutron source like ^{252}Cf is employed to induce fissions in ^{235}U. However, in cases where a thermal neutron source is not readily available, an Am-Be source, known for emitting fast neutrons, is utilized for fission induction. This approach is adopted primarily due to the unavailability of a thermal neutron source. In our case, the primary goal was to induce fission events that produce distinguishable traces on Solid State Nuclear Track Detectors (SSNTD). The approach employed to generate these fission events is notably straightforward and involves minimal experimental complexity.

Results

In this section, we present the results of our study, including a comparison of simulated tracks generated by the Fission Track Analysis Trainer Application (FTA Trainer) with 'real data' obtained from experiments. Our investigation involved assessing the fidelity of simulated tracks in reproducing actual fission track characteristics, with a focus on track width analysis, the influence of various parameters on fission track appearance, and exploring applications of the FTA Trainer software. Figure 4. displays a side-by-side comparison of simulated and real fission tracks, demonstrating remarkable visual congruence. This comparison reveals that the FTA Trainer captures key characteristics of real fission tracks, validating the reliability of our simulations. Notably, this fidelity was achieved through a process of iterative refinement, where initial simulations were compared to real fission clusters. Through a series of adjustments and fine-tuning, we progressively converged upon a closer resemblance between simulated fission cluster images and their real counterparts. The FTA Trainer software can generate a wide variety of fission track clusters, as determined by the user. Examples illustrated in Fig. 6.

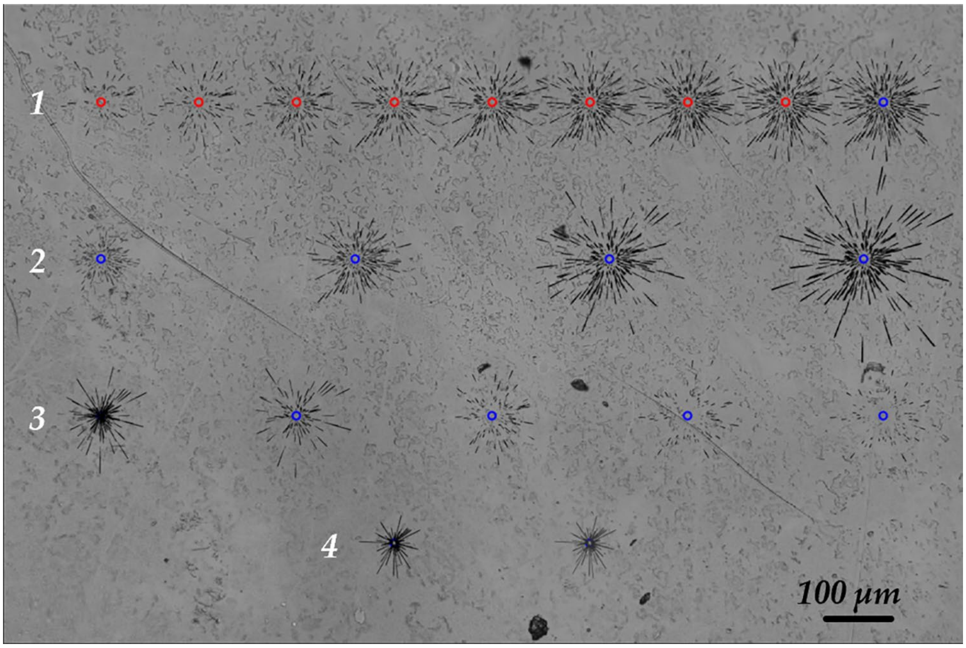


Fig. 6 Demonstration of the clusters plotting versatility in the FTA Trainer application

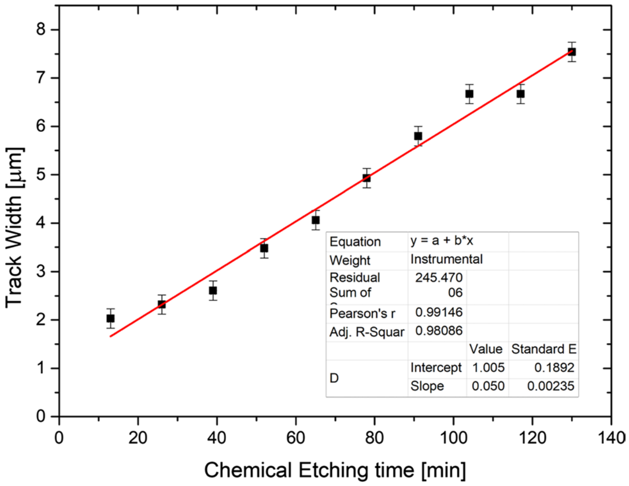


Fig. 7 Linear relation between Track width and chemical etching time

In Fig. 6, Row No.1 displays the option of adding a number of tracks. Row No.2 displays the option of increasing the diameter of the whole cluster. Row No.3 displays the influence of different depths of a fissile particle in on the simulated tracks in LEXAN® detector and Row No.4 displays the option of changing the color of the clusters in a grey scale. By optimizing the configuration of the parameters, it is easy to replicate many complex formations of clusters.

The analysis of actual fission track width with respect to chemical etching time demonstrated a linear dependence as shown in Fig. 7. Below 13 min, no tracks were identified.

Figure 8 illustrates four simulations conducted with the FTA Trainer, emphasizing the linear correlation and showcasing the progressive expansion of track width in relation to etching time."

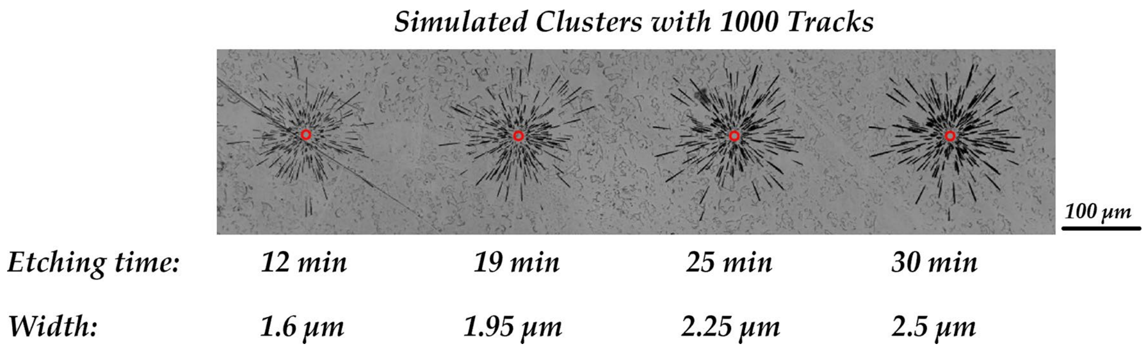


Fig. 8 Single Clusters, generated using the FTA Trainer application, with different etching times, showing the effect of varying etching times on the track width

The FTA Trainer software application utilizes practical observations, allowing users to have greater flexibility in selecting etching times for generating fission tracks, which, in turn, enhances the authenticity of these tracks.

The Assessment tool of the FTA Trainer can create training simulations for operators to improve their judgment and accuracy to cut suspected areas of the "catcher", which consists fissile particles, before working on real samples. In addition, this tool can be used for practice and recertification. Figure 9 shows test progress, whether the operator selects rectangular regions of interest (ROIs).

It's possible to visually compare the selected areas to the generated rectangles. The comparison provides a measure of the user's judgment (Fig. 10) and a total score.

Using the FTA Trainer, we investigated the track length distribution of various databases simulated in GEANT4 to understand the dependence of fission track lengths in different sample compositions. First, we simulated ^{235}U, see Fig. 11.

Next, we simulated fission track lengths as a function of the presence of soil in the sample (the particle size was 1 μm diameter and contained a mixture of soil with ^{235}U), as

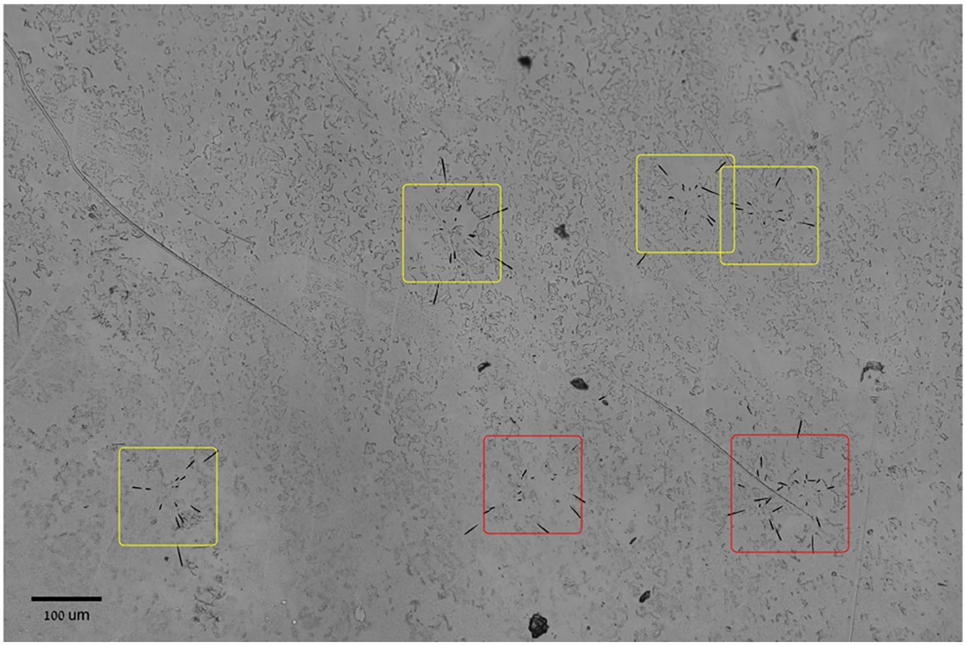


Fig. 9 Manual Cutting Area Test in progress. The operator chooses cut positions without overlapping (in yellow) and those with overlapping (in red)

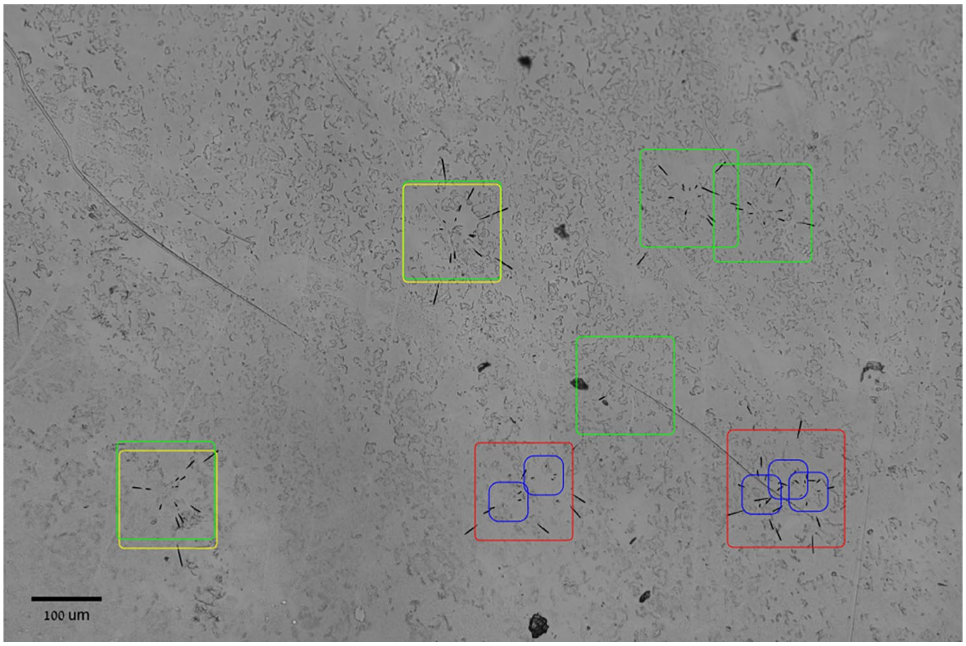
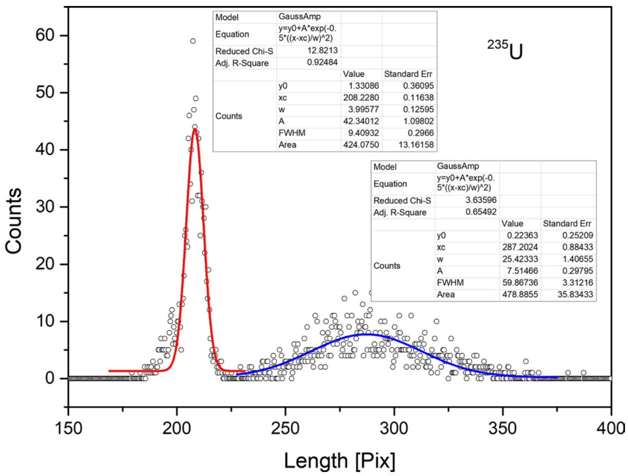


Fig. 10 Comparison between the suggested cut positions by the operator to the ideal cut positions which are calculated by the software. Green rectangles present POIs without overlapping, and blue rectangles present POIs with overlapping



Model	GaussAmp		
Equation	y=y0+A*exp(-0.5*((x-xc)/w)^2)		
Reduced Chi-S	12.8213		
Adj. R-Square	0.92484		
		Value	Standard Err
Counts	y0	1.33086	0.36095
	xc	208.2280	0.11638
	w	3.99577	0.12595
	A	42.34012	1.09802
	FWHM	9.40932	0.2966
	Area	424.0750	13.16158

Model	GaussAmp		
Equation	y=y0+A*exp(-0.5*((x-xc)/w)^2)		
Reduced Chi-S	3.63596		
Adj. R-Square	0.65492		
		Value	Standard Err
Counts	y0	0.22363	0.25209
	xc	287.2024	0.88433
	w	25.42333	1.40655
	A	7.51466	0.29795
	FWHM	59.86736	3.31216
	Area	478.8855	35.83433

Fig. 11 ^{235}U Track lengths Gaussian histograms of low and high energy fission products for 2000 fission incidents

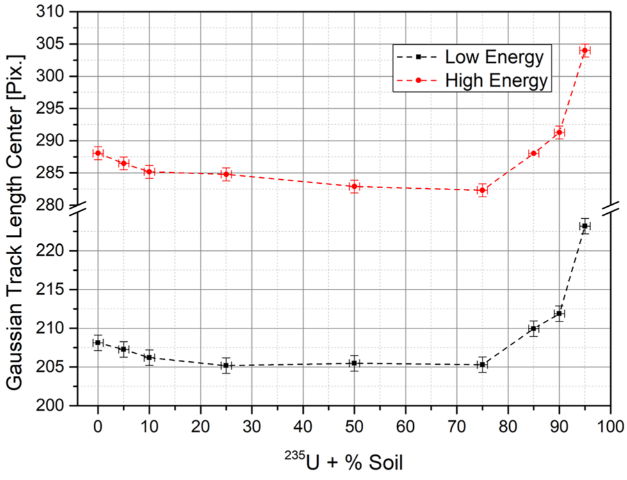


Fig. 12 ^{235}U Track lengths Gaussian histograms of low and high energy fission products as a function of soil concentration in a sample with 1 µm diameter

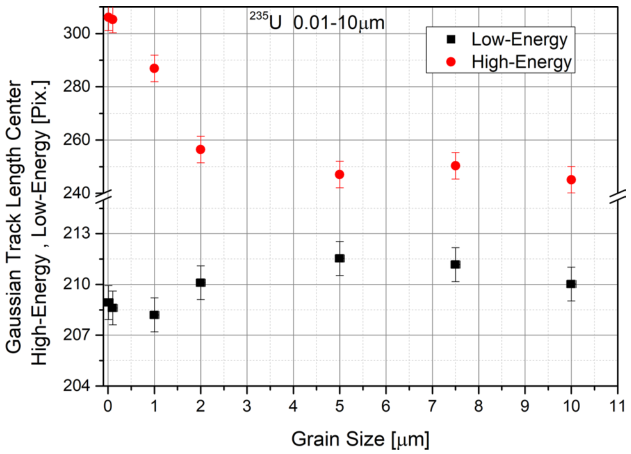
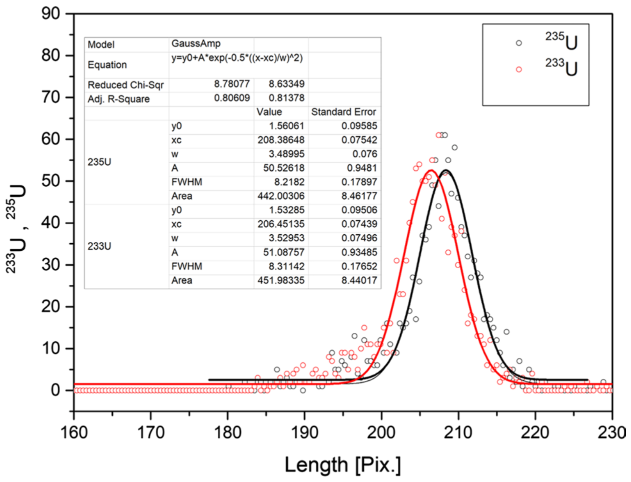


Fig. 13 ^{235}U Track lengths Gaussian histograms of low and high energy fission products as a function of increasing particle size



Model	GaussAmp		
Equation	y=y0+A*exp(-0.5*((x-xc)/w)^2)		
Reduced Chi-Sqr	8.78077	8.63349	
Adj. R-Square	0.80609	0.81378	
		Value	Standard Error
235U	y0	1.56061	0.09585
	xc	208.38648	0.07542
	w	3.48995	0.076
	A	50.52618	0.9481
	FWHM	8.2182	0.17897
	Area	442.00306	8.46177
233U	y0	1.53285	0.09506
	xc	206.45135	0.07439
	w	3.52953	0.07496
	A	51.08757	0.93485
	FWHM	8.31142	0.17652
	Area	451.98335	8.44017

Fig. 14 Difference between the distribution of the Gaussian histograms of ^{235}U and ^{233}U at low energy fission products

shown in Fig. 12. The enhancement of the lower and upper regions in the graph are due to the average density of the model and its composition.

The effect of grain size was checked for particle sizes between 0.01–10 µm because the heavy absorption in the ^{235}U is significant. Figure 13 indicates that the Gaussian histogram peaks approach each other as a function of increasing particle size.

Fissile isotopes yield unique fission products. Figure 14 shows a significant difference between the distribution of the Gaussian histograms of ^{235}U and ^{233}U at low energy fission products.

Furthermore, upon closer examination, it becomes apparent that the image data initially possessed two dimensions, representing the projection of the fission tracks. However, it's worth noting that our Solid-State Nuclear Track Detector (SSNTD) is transparent [5, 14], enabling us to extend this imagery into a three-dimensional realm through a process of deep auto-focusing known as Z-Stack imaging. This specialized approach utilizes a fluorescence agent known as ARDROX® 970 P23. By compiling an array of these precisely focused images into a stack, we were able to achieve a tomographic-style visualization of the sample. The result can be seen in Fig. 15, which highlights the fluorescent diffractogram, emphasizing the fission tracks while effectively eliminating extraneous artifacts typically associated with noise.

Figure 16 demonstrates image data obtained from the ECHO microscope. The upper image has the focusing depth encoded in pseudo-color. The lower image is a superposition

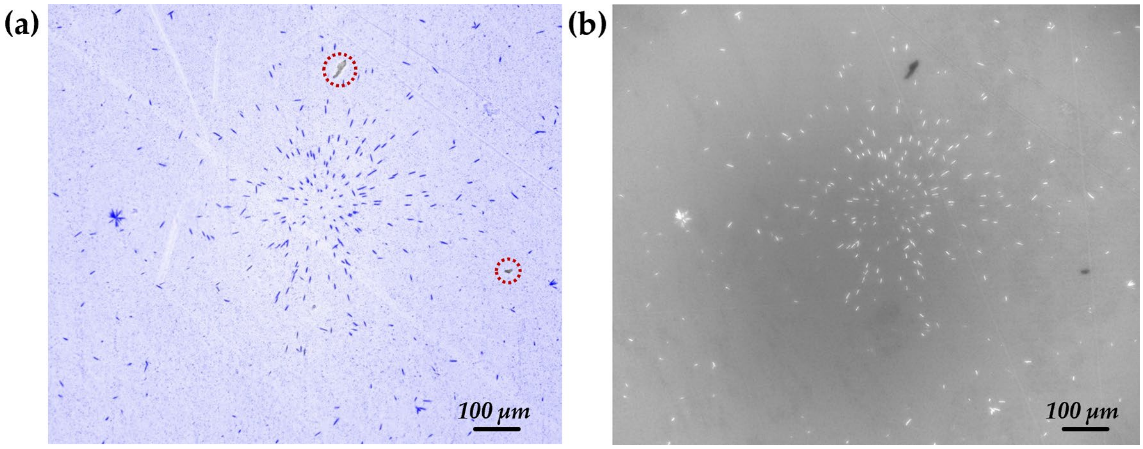


Fig. 15 Microscope fluorescent diffractograms of real fission tracks cluster, **a** fluorescent filter, and **b** bright field. The red circles indicate noise artifacts

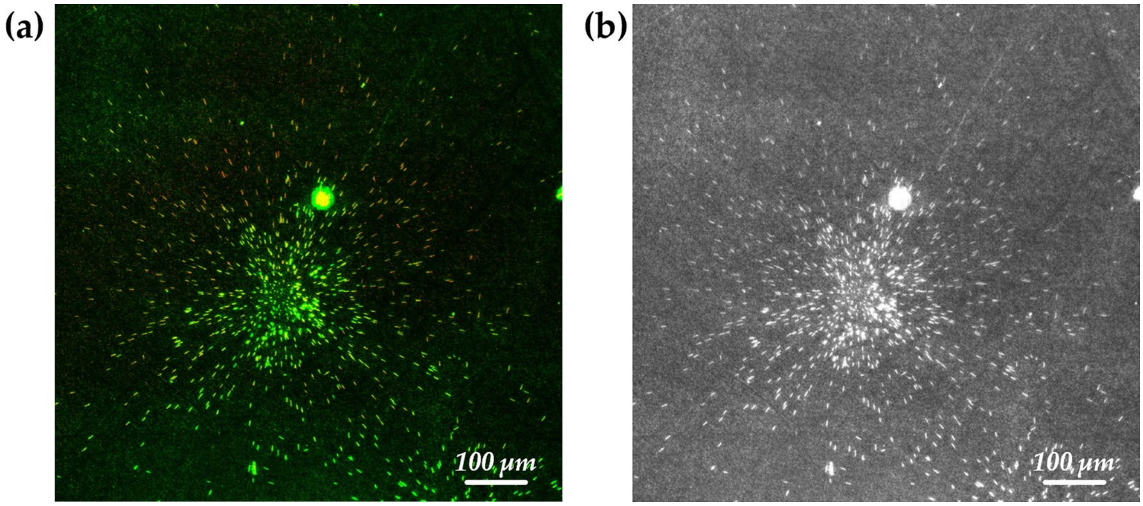


Fig. 16 Image data obtained from the ECHO microscope with a fluorescence agent ARDROX® 970 P23. **a** fluorescent filter and **b** bright field

of Z-Stack images (Merge of multiple depths); the SNR is not improved because of adding different focuses in the stack. The Set-up described in Appendix E.

Discussion

The FTA method belongs to the particle analysis approach. Therefore, in real samples, overlapping fission tracks make it challenging to analyze two particles individually, see Fig. 17a. The complexity is because of the limitation of cutting the size of the ROI in the sample. These incidents should be avoided because they cause large deviation in the results of the particle enrichment due to sample averaging. Sometimes real samples obtain clusters of the same size, but the enrichment of the fissile material and each particle mass are different. Figure 17b demonstrates a simulation for this kind of scenario.

The app can take into consideration the cross-section of the material affected by the incident neutron energy. The thermal neutron fission cross section of ^{235}U is ~ 583 barns [15]. For fast neutrons, the fission cross section drops to 1 ÷ 10. An example is shown in Fig. 17c, d.

The FTA Trainer can generate simulation that gives a visual demonstration of the ideal estimated rectangle size needed for a cutting suspected sample area before sending it to further particle analysis (i.e., ICPMS). Clusters that overlap are distinguished by a nearness criterion threshold.

If a sample contains large quantities of natural U particles with high mass, it will be hard to find low-mass enriched particles using FTA. In this case, if the difference in the enrichments is high enough, it can be detected with the

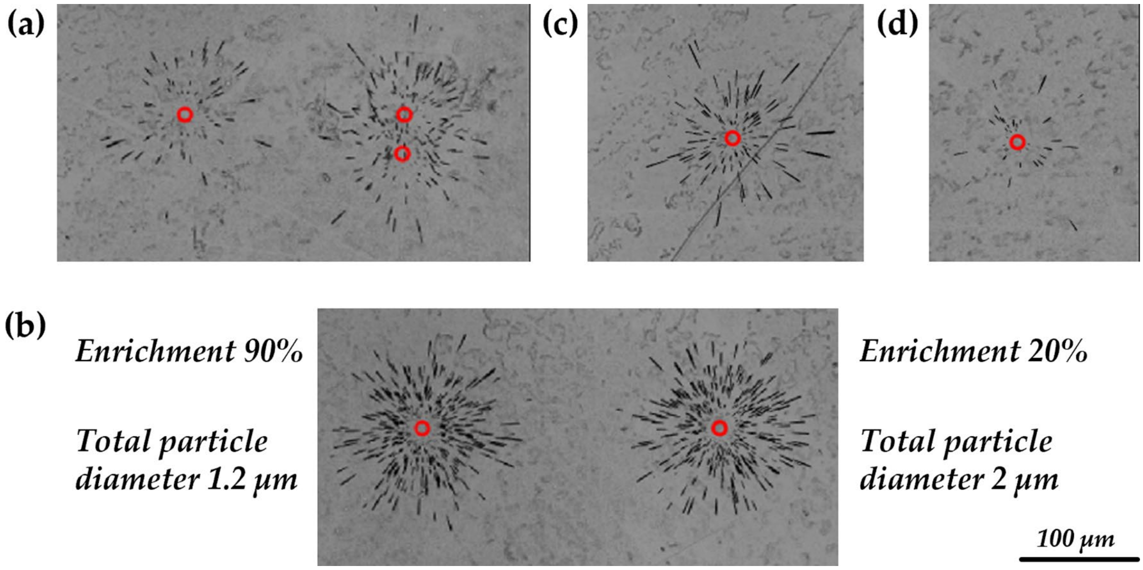


Fig. 17 **a** Mixed clusters **b** Similar clusters with different generating parameters **c** Poor Cluster as a result of Fast neutron Flux with a cross section of 10 barns **d** Rich Cluster as a result of Thermal neutron Flux with a cross section of 583 barns

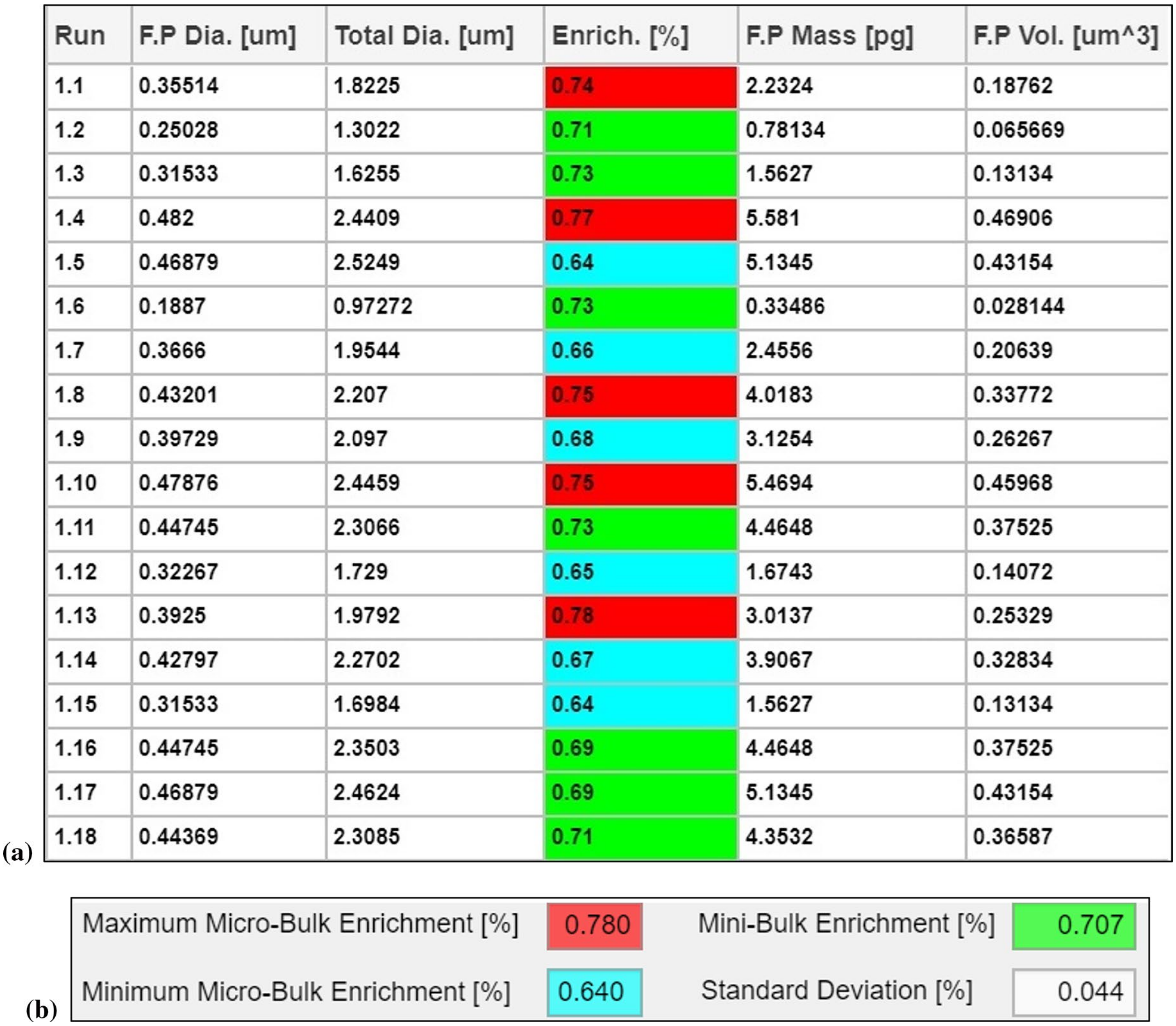
mini-bulk analysis using ICPMS. The presence of enriched or depleted U particles is a good indication for identifying if the sample is a by-product of a nuclear process. When a sample contains a variety of enriched particles, only micro bulk analysis using ICPMS can detect the true diverse enrichments. Otherwise, the particles may be detected with wrong enrichment (due to enrichment averaging). Using the FTA Trainer application, it is straightforward to demonstrate that statement. Figure 18 shows a batch of 18 simulations of U particles with different characteristics. The minimal micro bulk enrichment is 0.64% (Depleted), and the maximal micro bulk enrichment is 0.78% (Enriched), although the mini-bulk enrichment is 0.707% (Natural enrichment). Note: 'F.P' Stands for fissile particle, 'Dia.' stands for diameter, 'Enrich.' Stands for enrichment and 'Vol.' Stands for volume.

To avoid over lapped clusters, it is essential to estimate the correct amount of the dissolved material containing the test particles in the LEXAN® foil inserted between two SSNTD detectors in the "sandwich formation". Too many test particles cannot be cut separately (for the ICPMS measurement). There are cases when the estimation is correct, but still, the dissolved piece of LEXAN® contains two or more close particles. In this case, the particles area in the foil can be cut into small new pieces and dissolved again. This action gives the option to separate the particles. If a real sample contains overlapped particles, the analysts must cut a new model from the given foil and restart the process to avoid deviation in the results. By using the mass calculation option in the FTA Trainer application, it's possible to develop a method that can help estimating the correct amount of dissolved material to avoid nearby particles in micro bulk ICPMS analysis. The fissile mass calculator in Log tab, as shown in Fig. 18b, gives an example of a simulation. With enough simulation statistics, a right amount of material can be defined before preparing a sample for analysis.

Conclusions

In this study, we have presented a comprehensive exploration of fission track analysis (FTA) in the context of nuclear forensics. FTA is a powerful technique used for identifying fissile materials, an essential component of nuclear forensics and safeguards investigations. The critical challenge we aimed to address was the manual and operator-dependent nature of FTA, which can lead to issues such as overlapping tracks, potentially affecting the accuracy of results.

The central focus of our work has been the development of the FTA Trainer Application, a novel software tool. This application allows accurate modeling of fission track clusters on Solid State Nuclear Track Detectors (SSNTD),



Run	F.P Dia. [um]	Total Dia. [um]	Enrich. [%]	F.P Mass [pg]	F.P Vol. [um^3]
1.1	0.35514	1.8225	0.74	2.2324	0.18762
1.2	0.25028	1.3022	0.71	0.78134	0.065669
1.3	0.31533	1.6255	0.73	1.5627	0.13134
1.4	0.482	2.4409	0.77	5.581	0.46906
1.5	0.46879	2.5249	0.64	5.1345	0.43154
1.6	0.1887	0.97272	0.73	0.33486	0.028144
1.7	0.3666	1.9544	0.66	2.4556	0.20639
1.8	0.43201	2.207	0.75	4.0183	0.33772
1.9	0.39729	2.097	0.68	3.1254	0.26267
1.10	0.47876	2.4459	0.75	5.4694	0.45968
1.11	0.44745	2.3066	0.73	4.4648	0.37525
1.12	0.32267	1.729	0.65	1.6743	0.14072
1.13	0.3925	1.9792	0.78	3.0137	0.25329
1.14	0.42797	2.2702	0.67	3.9067	0.32834
1.15	0.31533	1.6984	0.64	1.5627	0.13134
1.16	0.44745	2.3503	0.69	4.4648	0.37525
1.17	0.46879	2.4624	0.69	5.1345	0.43154
1.18	0.44369	2.3085	0.71	4.3532	0.36587

Maximum Micro-Bulk Enrichment [%] 0.780
Mini-Bulk Enrichment [%] 0.707
Minimum Micro-Bulk Enrichment [%] 0.640
Standard Deviation [%] 0.044

Fig. 18 **a** Log of simulation with **b** calculated data showing that mini-bulk analysis has natural enrichment and forensic data concerning both enriched and depleted particles is absent due to averaging enrichments

offering various advantages. It serves as a valuable operator training and performance assessment tool, improving the proficiency of individuals engaged in cutting operations on Points of Interest (POIs). Moreover, it has the potential to transform traditional FTA into a more robust and reliable method, reducing the likelihood of errors resulting from manual procedures.

In our study, we have examined several key aspects:

- The linear dependence of single fission track width on chemical etching time, which holds significant practical implications for controlling the etching process when dealing with real-world samples.
- The influence of various parameters, such as the presence of soil in the sample, particle size, fissile isotope type, and energy distribution of fission products on the appearance of fission track clusters. These insights provide a foundation for distinguishing between different fissile isotopes, enhancing the capabilities of nuclear forensic analyses.

While our simulations are promising, we acknowledge the importance of validating our results with real-world data.

Our preliminary experiments involving comparisons with real FTA data indicate alignment between simulated and real data images. However, comprehensive, and in-depth comparisons will be an essential focus of future research.

In summary, our study has laid the groundwork for understanding fission track cluster behavior and its applications in nuclear forensics. The FTA Trainer Application offers enhanced reliability for human analysis in this field, with the anticipation that it may pave the way for further advancements in nuclear forensic investigations.

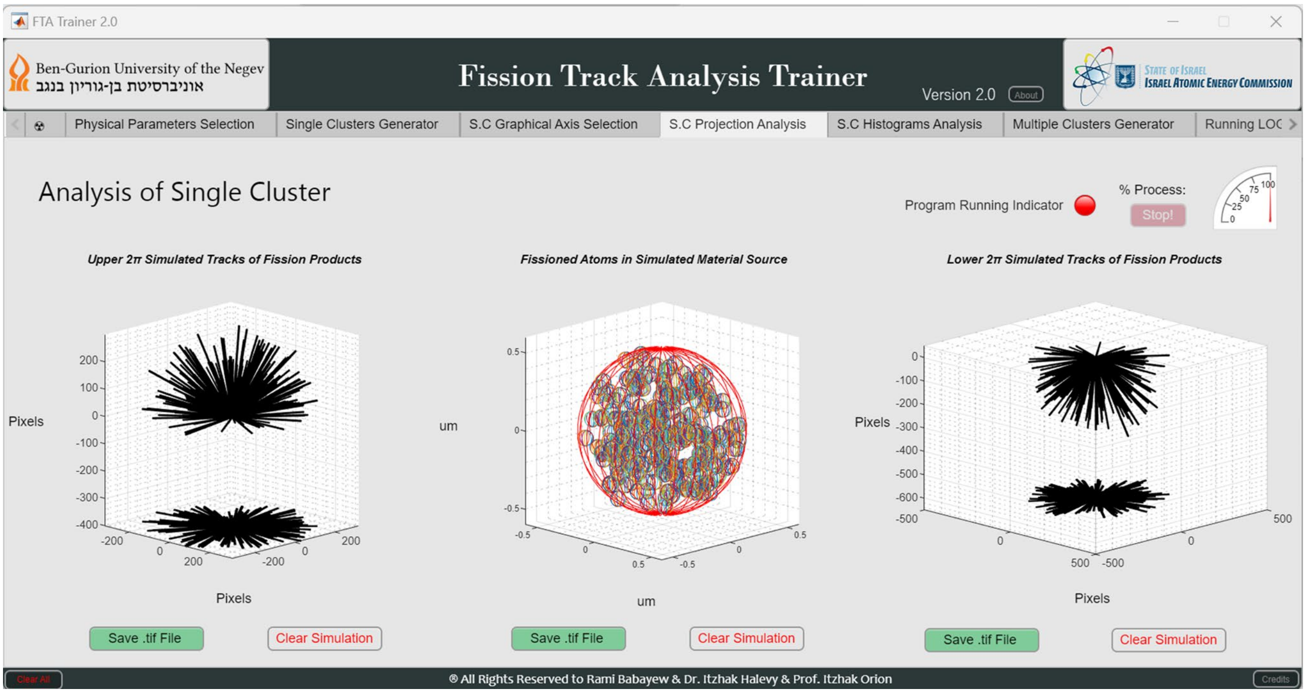
Appendix A

Minimum computer requirements:

1. Intel(R) Core (TM) i7-1065G7 CPU 1.50 GHz.
2. RAM: 16 GB.
3. Windows 10 operating system.
4. Free space memory: 5 GB.

Appendix B

FTA Trainer GUI Demonstration: 'Projection Analysis Tab' showing an example of simulated projection tracks on upper and lower SSNTDs (Left and right graphs respectively). In addition, the middle graph shows a 3D simulation of 3D which mapping the exact sites of fissions in the simulated particle (each fission mapped by colorful sphere, the red sphere is the entire fissile particle).



Appendix C:

FTA trainer software—main steps involved in the development process:

a) Data Collection and Input:
- Obtain a clear background image that serves as the background for the Fission Track model.
- Utilize a database created using GEANT4, which contains the trajectories of fission products. This database is essential for modeling the tracks.

b) User Input and Preferences:
- Receive user input to determine the specific characteristics of the Fission Track model they want. This could include parameters such as track density, track length, etch pit size, and other features.

c) Model Initialization:
- Create a blank canvas by overlaying the clear background image.
- Initialize the simulation environment with user-defined parameters and the physical data needed for the modeling process.

d) Track Generation:
- Use trigonometry and physics equations to calculate the positions and shapes of individual fission tracks.
- The physical data incorporated into the application, such as the energy of fission products, can be used to model the trajectories and interactions of these particles.

e) Rendering and Visual Representation:
- Overlay the generated fission tracks onto the clear background image.
- Apply appropriate coloring and visual effects to make the tracks resemble microscope images.

f) Quality Control and Validation:
- Implement mechanisms to ensure the accuracy and realism of the generated Fission Track models.
- Compare the output with expected results or experimental data to validate the accuracy of the simulation.

g) User Interaction and Feedback:
- Provide the user with the generated Fission Track model.
- Collect feedback from the user to make any necessary adjustments or refinements to the model.

h) Export and Reporting:
- Allow users to export the generated Fission Track model in various formats, such as images or data files, for further analysis or publication.

i) Documentation and Support:
- Provide comprehensive documentation to help users understand how to use the application effectively. User Guide for FTA Trainer Application: Link. Please note that the file is currently under construction and will be updated.
- Offer customer support or user assistance for any issues or questions that may arise during the modeling process.

j) Optimization and Performance:
- Implement performance optimizations to ensure efficient execution, especially if the application involves complex calculations or large datasets.

k) Security and Data Management:
- Ensure data security and proper management of the GEANT4 database and any user-generated content.

l) Future Development and Updates:
- Plan for future updates and enhancements to the application, including the incorporation of new research findings or user-requested features.

By following these key steps, the application can successfully replicate Fission Track models resembling microscope images.

2. FTA Trainer full capabilities list:

Generating fission tracks clusters by optimizing:

a) Size of a fissile particle.
b) Enrichment.
c) Mass of fissile particle.
d) Specific fissile isotope.
e) Neutron Flux.
f) Energy of neutrons used to radiate the particle.
g) Radiation duration.
h) Geometric Parameters.

Extra features for clarification of the simulated process and versatility:

i) Multi Cluster generator (arbitrary and systematic).
j) Different depths and slices of particle in the sample foil.
k) 3-Dimensional visualization of the fission clusters.
l) 3-Dimensional visualization of the fissions in the radiated particle.
m) Projection of upper and lower SSNTD detectors.
n) Nearness test for detecting potential overlapped clusters.
o) Etching time influence on clusters based on semi-empiric experiment.
p) ROI Cutting estimation ideal areas.
q) Operator certification test.

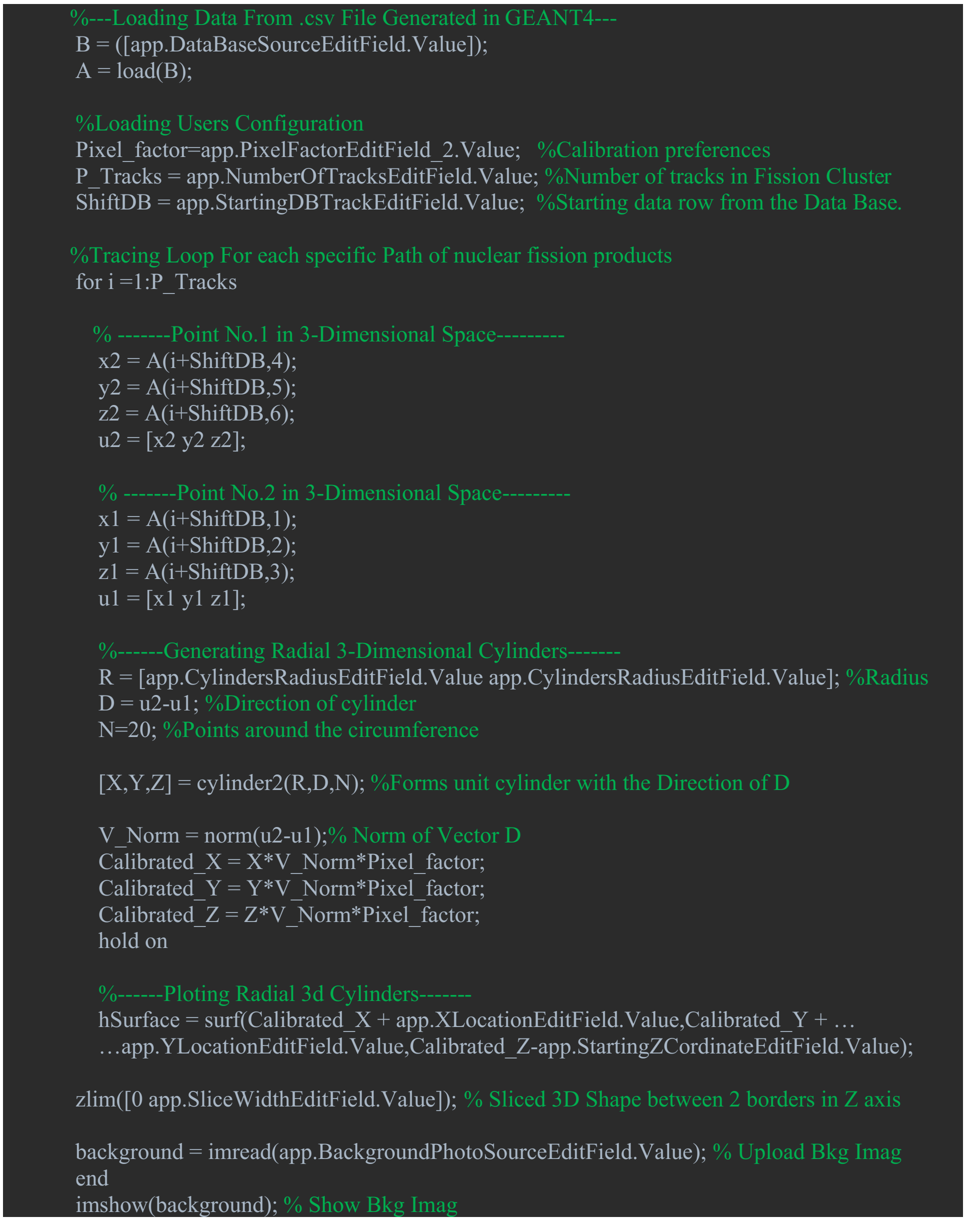
In addition, the following parameters can be Calculated:

a) Calculation of a number of tracks based on physical parameters.
b) Calculation of total fissile mass of a batch of clusters.
c) Mini bulk and Micro bulk enrichments.
d) Histograms of the fission products track lengths and the projected length (after 3-dimensional slicing).

e) Final score for tested operator (manual ROI cutting test).

3. FTA Trainer code example:

3.1. Part of a code which presents a segment dedicated to tracing the path of fission products, with a focus on generating cylinders and creating sliced 3D plots depicting the trajectory of fission products.



```
%---Loading Data From .csv File Generated in GEANT4---
 B = ([app.DataBaseSourceEditField.Value]);
 A = load(B);

 %Loading Users Configuration
 Pixel_factor=app.PixelFactorEditField_2.Value;   %Calibration preferences
 P_Tracks = app.NumberOfTracksEditField.Value; %Number of tracks in Fission Cluster
 ShiftDB = app.StartingDBTrackEditField.Value;  %Starting data row from the Data Base.

%Tracing Loop For each specific Path of nuclear fission products
 for i =1:P_Tracks

    % -------Point No.1 in 3-Dimensional Space---------
     x2 = A(i+ShiftDB,4);
     y2 = A(i+ShiftDB,5);
     z2 = A(i+ShiftDB,6);
     u2 = [x2 y2 z2];

     % -------Point No.2 in 3-Dimensional Space---------
     x1 = A(i+ShiftDB,1);
     y1 = A(i+ShiftDB,2);
     z1 = A(i+ShiftDB,3);
     u1 = [x1 y1 z1];

     %------Generating Radial 3-Dimensional Cylinders-------
     R = [app.CylindersRadiusEditField.Value app.CylindersRadiusEditField.Value]; %Radius
     D = u2-u1; %Direction of cylinder
     N=20; %Points around the circumference

     [X,Y,Z] = cylinder2(R,D,N); %Forms unit cylinder with the Direction of D

     V_Norm = norm(u2-u1);% Norm of Vector D
     Calibrated_X = X*V_Norm*Pixel_factor;
     Calibrated_Y = Y*V_Norm*Pixel_factor;
     Calibrated_Z = Z*V_Norm*Pixel_factor;
     hold on

     %------Ploting Radial 3d Cylinders-------
     hSurface = surf(Calibrated_X + app.XLocationEditField.Value,Calibrated_Y + …
     …app.YLocationEditField.Value,Calibrated_Z-app.StartingZCordinateEditField.Value);

 zlim([0 app.SliceWidthEditField.Value]); % Sliced 3D Shape between 2 borders in Z axis

 background = imread(app.BackgroundPhotoSourceEditField.Value); % Upload Bkg Imag
 end
 imshow(background); % Show Bkg Imag
```

Note: After slicing the 3D plot of the cylinder, the software provides the capability to observe the sliced object from an 'above' perspective, allowing users to view the plot in a 2D representation.

Appendix D

The microscope used for actual track width analysis is Nikon DSQI2 Y-TV55. Microscope Setups details:

1. Setup No. 1:
- Nikon S Plan Flour.
- ELWD 20X/0.45.
- ∞/0–2 WD 8.2 -6.9.
2. Setup No. 2:
- Nikon S Plan Flour.
- ELWD 40X/0.60.
- ∞/0–2 WD 3.6 -2.8.
3. Setup No. 3:
- Nikon N. Plan Apo λ.
- OFN25WD20.
- MRD000045
4. Setup No. 4:
- Nikon Tu plan Flour.
- ELWD 10X/0.30.
- ∞/0 EPI.

Appendix E

The microscope that was used for fluorescence analysis is ECHO REVOLUTION. Microscope Setups details:

1. Setup No. 1:
- OLYMPUS UIS2
- LUC Plan FLN.
- 20X/0.45 Ph1.
- ∞/0–2/FN22.
2. Setup No. 2:
- OLYMPUS UIS2
- U Plan FLN.
- 10X/0.30 Ph1.
- ∞/-/0–2/OFN26.5.
3. Setup No. 3:
- OLYMPUS UIS2
- U Plan FLN.
- 4X/0.13 PhL.
- ∞/-/OFN26.5.

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Data availability The data that support the findings of this study are available upon reasonable request. Due to confidentiality agreements and ethical considerations, certain sensitive or restricted data cannot be made openly accessible. Requests for access to these data may be directed to the corresponding author [ramibab@post.bgu.ac.il] or the institutional review board [halevy.itzhak.dr@gmail.com, iorion@bgu.ac.il] for further guidance on data access procedures.

Declarations

Conflict of interest The authors certify and declare that there are no conflicts of interest regarding the publication of this article.

Ethical approval This study was conducted in an objective and unbiased manner, and no external parties or organizations had any influence on the design, execution, or reporting of the research.

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