Radiation Physics and Chemistry

Elsevier LATEX templatel

--Manuscript Draft--

Manuscript Number:	
Article Type:	SI:Proceedings of ISRP-15
Keywords:	elsarticle.cls; LATEX; Elsevier; template
Corresponding Author:	Josip Batur, mag.phys. Ruder Boskovic Institute: Institut Ruder Boskovic CROATIA
First Author:	Josip Batur, mag.phys.
Order of Authors:	Josip Batur, mag.phys.
	Davorin Sudac
	Ilker Meric
	Helge E.S. Pettersen
	Vladivoj Valkovic
	Jasmina Obhodas
	Karlo Nad
	Zeljko Orlic
	Milivoj Uroic
	Marko Gacina
	Adaleta Perkovic
Abstract:	This template helps you to create a properly formatted LATEX manuscript.
Suggested Reviewers:	Giancarlo Nebbia giancarlo.nebbia@pd.infn.it
	Bertrand Perot bertrand.perot@cea.fr
	Cedric Carasco cedric.carasco@cea.fr

RESEARCH HIGHLIGHTS

Temporal and spatial resolution of the neutron probe for C/O logging with associated alpha particle technique was measured. PET bottle filled with diesel was used as a target for inelastic neutron scattering. Measurements were supplemented with Monte Carlo simulations of the experiment using MCNP6.2. Temporal resolution was found to be 2 ns, with corresponding spatial resolution of 10 cm.

Click here to view linked References

Temporal and spatial resolution of the neutron probe for C/O nuclear well logging

J.Batur[a], D.Sudac[a], V.Valkovic[a], I.Meric[b], H.E.S.Pettersen[c], K.Nad[a], M.Uroic[a], Z.Orlic[a], A.Perkovic[d], M.Gacina[d], J.Obhodas[a]

[a] *Rudjer Boskovic Institute, Bijenicka c.54, 10000 Zagreb, Croatia*
[b] *Department of Computer Science, Electrical Engineering and Mathematical Sciences, Western Norway University of Applied Sciences, 5020 Bergen, Norway*
[c] *Department of Oncology and Medical Physics, Haukeland University Hospital, 5021 Bergen, Norway*
[d] *Laboratory for Rock and Fluid Analysis, INA-Industrija nafte d.d., Avenija Ve ́ceslava Holjevca 10, 10020 Zagreb, Croatia*

Abstract

The potential of detecting particular formation elements inside boreholes, separated by small distances, was investigated. Neutron probe, designed for C/O nuclear well logging and with associated alpha particle method (AAP) as its operational principle, was used. The measurements consisted of irradiating 0.5 l diesel-filled plastic bottle with fast tagged neutron beam. The bottle was placed in four positions along the axis of the probe, spaced by $\sim$ 10 cm; time spectrum for each position was obtained. The peaks in time spectra are separated by $\sim$ 2 ns, suggesting that neutron probe with AAP can, in principle, distinguish between parts of the formation separated by 10 cm. The results were confirmed by Monte Carlo (MC) simulations of the experiment.

Keywords: `Tagged neutrons`, Monte-Carlo simulations, Temporal resolution, Spatial resolution

---

Introduction

Conventional logging identification, evaluation and interpretation of thin-bedded reservoirs sometimes could result in a difference between the prediction and actual reservoir production, especially in cases where no core material is available. Locating the depth intervals of production zones could be challenging and might lead to underestimated reserves. Improving the identification and evaluation is of great importance in a segment of reservoir development. For example, the gamma-ray log resolution for thin-bedded layers of sandstone reservoirs is low. Precise and accurate detection of lithology changes is possible if the core material is available. In cases of oil layers surrounded with mudstone rocks, the logging response will be affected by the surrounding rocks. That can give, for example, overestimated values of water saturation [1]. Even in a case where the core material is available, sometimes the validation of correlating log and core integration data does not ensure the exact petrophysical evaluations, especially quantitative determination of shale volume [2]. An additional challenge is also

---

*Corresponding author
Email address: `jbatur@irb.hr` (J.Batur)

a high-temperature environment which reduces the number of available technologies and at the same time increases the safety hazards and the cost of onsite procedures, especially within high-temperature geothermal reservoirs [3]

Neutron probes (NPs) designed for nuclear well logging have been proven to be a very efficient method of investigating hydrocarbon saturation of formations outside the borehole pipes (casing) [4, 5]. Its main components are usually a source of fast neutrons and one or two gamma and neutron detectors, all placed inside long and hollow metallic cylinder. Probe is lowered through the borehole pipe at steady rate as to analyze data at different conditions for extra information.

This type of neutron tool has been developed and used for decades, yielding numerous different methods for testing the formations outside the borehole casing. Since the thermal neutron capture cross section for chlorine is significantly larger than for carbon, hydrogen and oxygen, high-salinity oilfields were commonly investigated using the die-away method, which measures the lifetime of thermal neutrons absorbed in the formation of interest. Thermal neutron capture cross sections are comparable for oxygen, hydrogen and carbon and therefore, the die-away method is not suitable for investigating freshwater oilfields. To estimate the oil saturation in freshwater conditions, carbon-to oxygen (C/O) well logging is commonly used[6, 7]. Furthermore, C/O logging is convenient for examining rock formations that contain carbon[8]. The C/O method utilizes fast neutrons produced by a pulsed neutron source which excites the nuclei of chemical elements of which the formation is composed. Prompt gamma rays emitted through de-excitation of nuclei are detected by gamma detectors attached to appropriate electronics and subsequently analyzed. Pulsed neutron sources can also be used for geochemical elemental logging and neutron-gamma density logging [9, 10].

The main disadvantage of conventional NPs that utilize fast neutron activation analysis (FNAA) is significant background influence. In contrast, NPs that utilize the associated alpha particle technique (AAP), proposed by Valkovic et al.[11], were proven useful in detection of hidden explosives [12] and were shown to reduce background noise in C/O logging[13]. Namely, tagging neutrons with alpha particles enables detection of only those formations which are placed inside the tagged neutron cone. A neutron tool utilizing the AAP method was recently successfully tested in high-temperature environments (up to 175°C) with pulse amplitude degradation observed as the temperature was increased [14].

In this research, a temporal and spatial resolutions of 2 ns and 10 cm, respectively, estimated by [14] were proven experimentally and supplemented with Monte Carlo simulations of the measurements. The obtained results have a potential of improving the quality of borehole research even further.

Methods

Experimental setup and measurements

The AAP method is based on electronic collimation of the 14 MeV neutron beam, produced in $d+^{3}H \longrightarrow n+^{4}H$ reaction. Neutrons inelastically scatter off various nuclei that compose the target. Characteristic gamma rays, emitted through de-excitation of those nuclei were used as a start signal for time-of-flight (t-o-f) measurement. Alpha particles, on the other hand, were used as delayed stop signal.

The AAP probe is shown schematically in Fig.1. A tritium target with the active area of 6 mm in diameter is placed into the holder is being hit by the deuteron beam, inducing $^{3}\mathrm{H}(d,n)^{4}\mathrm{He}$ fusion reaction. Produced alpha particles were detected by YAP:Ce alpha detection scintillator 0.5 mm thick and 38 mm in diameter, with four equal optically divided parts. Scintillator is connected to the Hamamatsu R4177-01 photomultiplier tubes [14].

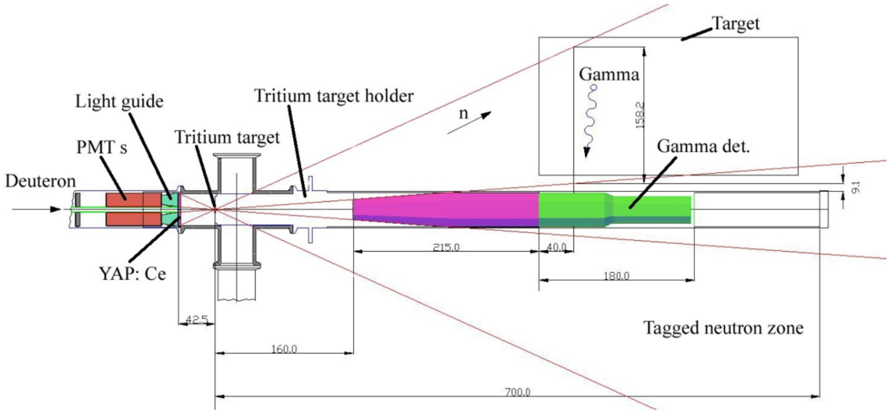

Figure 1: Scheme of a neutron probe for the AAP technique (units in mm). The target is shown placed inside the tagged cone of neutrons. Characteristic gamma rays emitted from the target are used as a start signal in time-of-flight measurements. The cone of alpha particles is shown on the left of the tritium target and is used as a stop signal.[14]

Characteristic gamma rays emmited from the target are detected by $\mathrm{LaBr_3}$:Ce scintillator, which showed satisfying characteristics when applied to the borehole research. Lead shielding in front of the $\mathrm{LaBr_3}$ provides protection from the background gamma rays produced by neutron interactions with probe casing, target holder etc.

It is worth noting (Fig.1) that for t-o-f measurement to be successful, one needs to place the target inside the tagged neutron cone, defined by the geometries and positions of the tritium target and YAP:Ce alpha scintillator.

The experimental setup is shown in Fig.2. A 0.5 l PET bottle filled with diesel was used as a target. The bottle was placed in four different positions separated by 10 cm and parallel to the axis of the probe and the measurements included time spectra and gamma-ray spectra.

Monte Carlo code

The experiment was simulated using MCNP6.2 (Monte Carlo n-Particle Transport Code)[15]. Studies using Monte Carlo have seen success in simulating characteristic gamma-ray neutron activation analysis of multiphase gas/water/oil/salt flow amounts in a deep sea [16, 17]. More recently, it confirmed significant background reduction in measuring C/O values using AAP [13].

Each simulation setup recreated the experimental geometry using basic MCNP6.2 structures, continuous cell volumes bounded by well defined surfaces. Setup was simplified (Fig.3.) and contained neutron source, lead shield, $\mathrm{LaBr_3}$ detector and diesel target. Each material that fills appropriate cell volumes were described using the material density and mass fractions of the elements that those materials were composed of.

Figure 2: Experimental setup. The PET bottle filled with diesel fuel is placed in one of the four positions for which the time spectra was measured. Instrumentation (scintillators, photomultipliers) is placed inside metallic cylinder.

For diesel fuel, an average chemical formula $C_{12}H_{23}$ was assumed and its mass fractions were calculated.

The point source emits 14 MeV neutrons in a narrow cone directed towards the diesel target. Each of the successive positions was spaced 10 cm in the direction of probe-axis (Fig.3.). Interactions of neutrons with the nuclei that constitute the materials were defined in input file by continuous-energy neutron cross-section libraries. MCNP6.2 uses the most recent default libraries ENDF/B-VII.1 for neutrons and ENDF/B-VI.8 for photons. Default data libraries used include gamma ray production for each element, with the exception of La-139.

Neutrons and photons were the only particles being tracked and default settings for neutron and photon physics were used in each simulation. Neutrons and photons were tracked down to 0 keV and 1 keV, respectively. Total of 2×10^9 particle histories were generated per simulation.

The time tally data was generated using the pulse-height tally card `f8`, time bin card T and energy deposition card `f6`. Specifically, `f8` tally is allowed with T card, but only in conjunction with special treatment tally card FT PHL. `F6` tally converts energy deposition to light equivalent and is used, together with FT PHL option, to subdivide the pulse-height tally. Finally, the pulse-height time tallies were generated and divided into 0.1 ns bins.

Results

Experimental results

Time spectra and prompt gamma-ray energy spectra for 0.5 l plastic bottle filled with diesel fuel placed on four different positions were measured. The first position is closest to the source, and the fourth position is farthest. Time spectra were shown in Fig.4. Peak shifts to the left in time spectrum (time window I) were clearly visible as

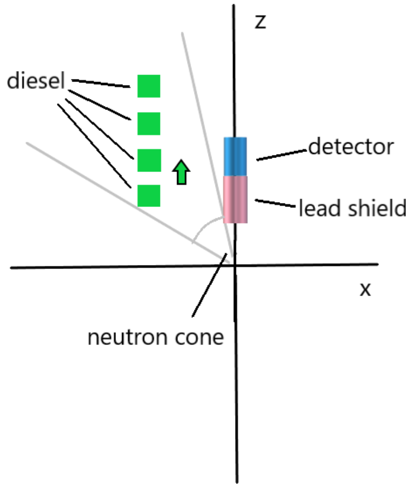

Figure 3: 2D scheme of simulated experimental setup. Point neutron source is placed at the origin. The arrow indicates movement of the PET bottle filled with diesel from the first position (closest to the neutron source) to the fourth position (farthest from the neutron source).

the target moves along the NP axis. Explaining time window II will require further testing. It is possibly due to the source neutrons penetrating through the lead shield and/or being scattered off diesel target.

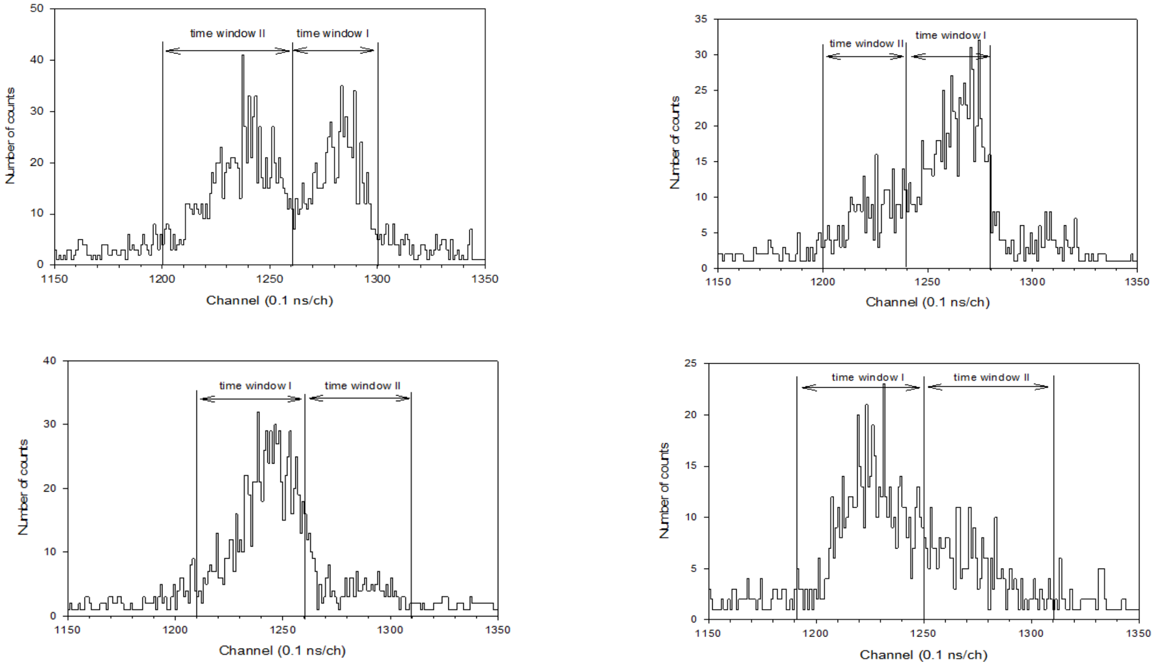

Figure 4: Time spectra for the first (top left), second (top right), third (bottom left) and fourth (bottom right) position of the diesel bottle. Time window I corresponds with prompt gamma rays and it shifts to the left for about 2 ns for each position. Time window II is possibly due to detection of source neutrons and scattered neutrons.

Peak shifts to the left as the target is moved away from the source is expected since

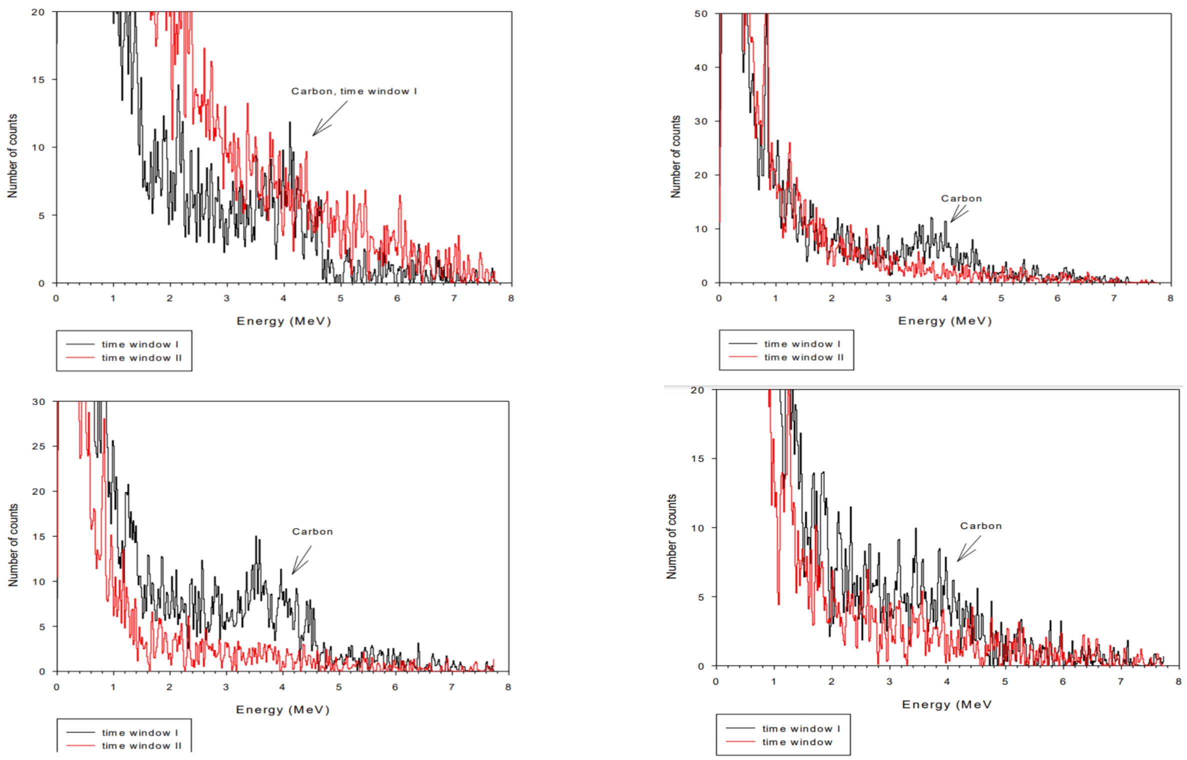

Figure 5: Prompt gamma-ray energy spectra for the first (top left), the second (top right), the third (bottom left) and the fourth (bottom right) position of the diesel bottle.

the prompt gamma rays are used as starting signal for the time-to-amplitude converter (TAC). Means of the time window I are shifted (2.2 ± 0.1) ns (between the first and the second position), (1.5 ± 0.1) ns (between the second and the third position) and (1.8 ± 0.1) ns (between the third and the fourth position). Errors of 0.1 ns are estimated using the fact that time spectra is collected in 0.1 ns bins. Subsequently, distances between adjacent positions are calculated using 5.2 cm/ns as the 14 MeV neutron speed and their values are, respectively: 11.4 ± 0.5 cm, 7.8 ± 0.5 cm and 9.4 ± 0.5 cm.

In addition to time spectra, prompt gamma-ray energy spectra for successive positions are provided (Fig.5). Carbon peaks in the energy spectra coincide with time window I.

Simulations

Monte Carlo simulation results are shown in Fig.6. Unlike the experimental time spectra, simulation time peaks shifted to the right, since every source neutron started at time $t = 0$. Means of the time spectra are shifted by (2.0 ± 0.1) ns for each successive position.

An analogue of the time window II did not appear in the simulations. This was the expected result if we assume that the time window II in the experiment came from scattered source neutrons, since default data libraries for La-139 don't include gamma production. However, considerable amount of pulses are tallied around 0.6-1.0 sh (6-10 ns) (Fig.6.). This is due to gamma-rays produced by inelastic neutron scattering in lead shield.

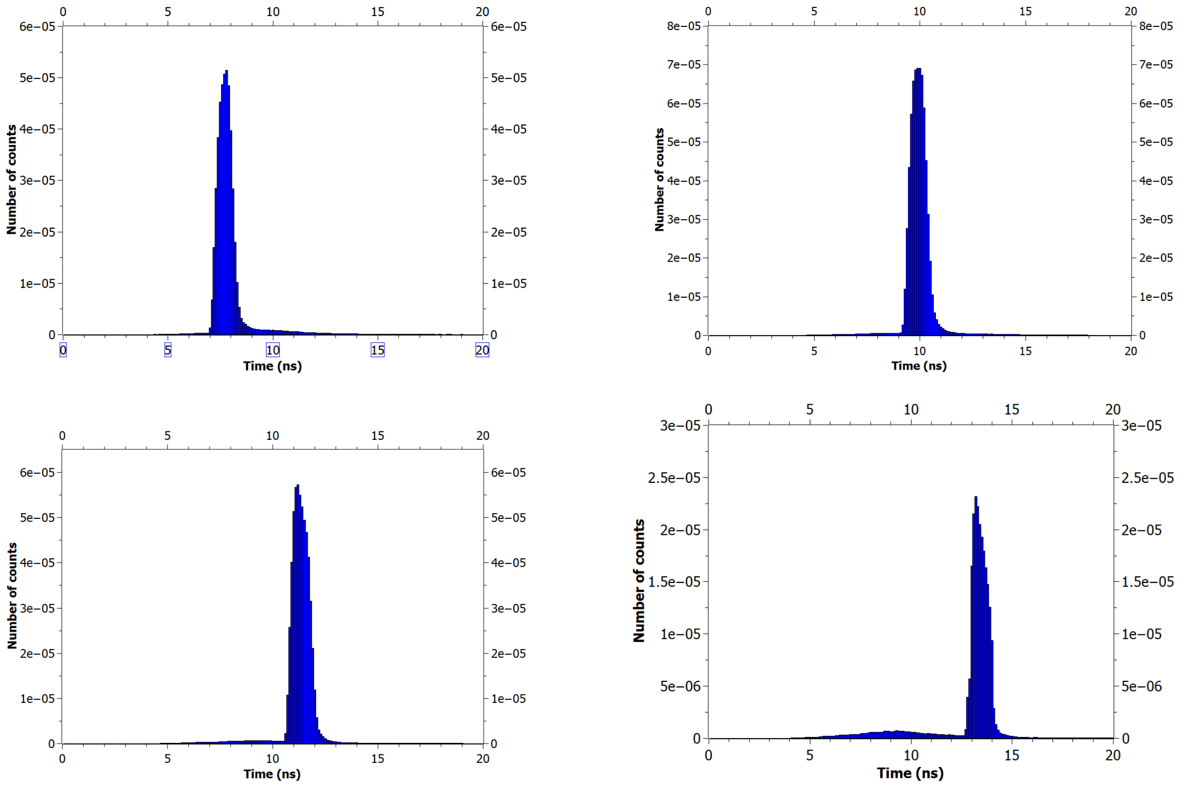

Figure 6: Time spectra generated with MCNP6.2 for different positions of diesel bottle is shown. Spectral peaks shift to the right by 0.2 sh (2 ns) for each successive position. Spectra for third and fourth position show pulse detection between 0.6 sh and 1.0 sh. Those pulses came from gamma-rays produced in the lead shield.

Conclusion

A neutron probe with the associated alpha particle technique as its operational principle has shown success in distinguishing small parts of the formation volume along the NP axis. Temporal resolution of such probe was shown to be at least 2 ns, allowing us, in principle, differentiating and measuring parts of the formation separated by 10 cm. Experimental results were supplemented and confirmed with MCNP6.2 simulations. Further research will try to explain pulses appearing as additional time window in time spectra. Our future endeavours will focus on more realistic simulations of associated alpha particle in borehole conditions.

Acknowledgments

This work has been fully supported by the Croatian Science Foundation under the project IP-2018-01-4060 “Advanced applications of 14 MeV neutrons”. The work of doctoral student Josip Batur has been fully supported by the “Young researchers’ career development project – training of doctoral students” of the Croatian Science Foundation, Croatia (DOK-2020-01). The work of H.E.S. Pettersen has been supported by the Trond Mohn Foundation (BFS2017TMT07).

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

References

1. Lu S, Deng R, Linghu S, Wu S. Identification and logging evaluation of poor reservoirs in x oilfield. *Open Geosciences* 2021;13(1):1013–27. URL: `https://doi.org/10.1515/geo-2020-0280`. doi:`doi:10.1515/geo-2020-0280`.
2. Yadav L, Dutta T, Sinha N. Reconciliation of Core and Log Data Analysis in Very Thin Reservoirs of Krishna Godavari Basin, India 2012;All Days. URL: `https://doi.org/10.2118/149017-MS`. doi:`10.2118/149017-MS`; sPE-149017-MS.
3. Serko S, Arthur G. Wireline evaluation technology in hpht wells 2005;All Days. URL: `https://doi.org/10.2118/97571-MS`. doi:`10.2118/97571-MS`; sPE-97571-MS.
4. Ellis D, Springer J. Well Logging for Earth Scientists. Springer; 2007. ISBN 978-1-4020-4602-5.
5. Valkovic V. 14 MeV Neutrons, Physics and Applications. CRC Press, Taylor & Francis Group; 2016. ISBN 13:978-1-4822-38006-6.
6. Al-Naggar TI, Abdalla AM, Harraz FA, Albargi HB. Alpha particles detection using p3ht conducting polymer-coated dam-adc. *Radiation Physics and Chemistry* 2022;190:109820. doi:`https://doi.org/10.1016/j.radphyschem.2021.109820`.
7. Schultz WE, Smith Jr HD. Laboratory and field evaluation of a carbon/oxygen (c/o) well logging system. *J Pet Technol, v 26, pp 1103-1110* 1974;URL: `https://www.osti.gov/biblio/4260131`. doi:`10.2118/4638-PA`.
8. Lawrence T. Continuous carbon/oxygen log interpretation techniques. *SPE 8366, 54th Annual Technical Conference and Exhibition* 1979;.
9. Wang , et al . A new method for calculating bulk density in pulsed neutron-gamma density logging. *Geophysics* 2020;85:1ND–Z30. doi:`10.1190/geo2018-0821.1`.
10. Wu W, Yue A, Xiang W, Wang S, Fan D. Influence of borehole and formation characteristics on elemental standard spectra in geochemical logging. *Journal of Geophysics and Engineering* 2017;14(5):1103–14. URL: `https://doi.org/10.1088/1742-2140/aa7304`. doi:`10.1088/1742-2140/aa7304`.
11. Valkovic V, et al . New neutron log for small scale variations in carbon/oxygen ratio 2015;62(2):407–14. doi:`10.1109/TNS.2015.2390671`.
12. Sudac D, et al . Inspections for contraband in a shipping container using fast neutrons and the associated alpha particle technique: Proof of principle. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* 2005;241:798–803. doi:`10.1016/j.nimb.2005.07.135`.

13. Sudac D, et al . C/o logging by using the associated alpha particle method: Proof of principle. *IEEE Transactions on Nuclear Science* 2022;doi:`10.1109/TNS.2021.3127578`; to be published.

14. Sudac D, et al . Development of a neutron probe for carbon-to-oxygen (c/o) oil logging equipped with a high-temperature alpha particle detector. *Fuel* 2021;:122843doi:`https://doi.org/10.1016/j.fuel.2021.122843`.

15. Werner C, et al . MCNP6.2 Release Notes. Los Alamos National Laboratory, report LA-UR-18-20808; 2018.

16. Wang J, Li F, Gardner RP. On the use of prompt gamma-ray neutron activation analysis for determining phase amounts in multiphase flow. *Measurement Science and Technology* 2008;19(9):094005. URL: `https://doi.org/10.1088/0957-0233/19/9/094005`. doi:`10.1088/0957-0233/19/9/094005`.

17. Meric I, Johansen GA, Holstad MB, Wang J, Gardner RP. Produced water characterization by prompt gamma-ray neutron activation analysis. *Measurement Science and Technology* 2011;22(12):125701. URL: `https://doi.org/10.1088/0957-0233/22/12/125701`. doi:`10.1088/0957-0233/22/12/125701`.

Declaration of interests

☒The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: