

Development of the Quality Assurance/Quality Control Procedures for a Neutron Interrogation System

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Abstract—In order to perform Quality Assurance/Quality Control (QA/QC) procedures for a system dedicated to the neutron interrogation of objects for the presence of threat materials one needs to perform measurements of reference materials (RM) i.e. simulants having the same (or similar) atomic ratios as real materials. It is well known that explosives, drugs, and various other benign materials, contain chemical elements such as hydrogen, oxygen, carbon and nitrogen in distinctly different quantities. For example, a high carbon-to-oxygen ratio (C/O) is characteristic of drugs. Explosives can be differentiated by measurement of both (C/O) and nitrogen-to-oxygen (N/O) ratios. The C/N ratio of the chemical warfare agents, coupled with the measurement of elements such as fluorine and phosphorus, clearly differentiate them from the conventional explosives. Here we present the RM preparation, calibration procedure and correlations attained between theoretical values and experimentally obtained results in laboratory conditions for C/O and N/C ratios of prepared hexogen (RDX), TNT, DLM2, TATP, cocaine, heroin, yperite, tetranitromethane, peroxide methylethylketone, nitromethane and ethyleneglycol dinitrate simulants. We have shown that analyses of the gamma ray spectra by using simple unfolding model developed for this purpose gave a nice agreement with the chemical formula of created simulants, thus the calibration quality was successfully tested.

Index Terms—.

I. INTRODUCTION

THE purpose of container inspection is to compare container cargo with its cargo manifest. Any deviation observed requires further investigation. Container inspection operations are strongly affected by the normal port operations. To this end, it is worthwhile saying that current standards and norms help in identifying suspected containers even before their arrival in the port of destination. However, the inspection of a suspected container can be done only when the container becomes available in the port area, e.g. unloaded from the ship and positioned according to the security officers' request at a specific location in the yard area. The suspected container can be inspected either manually or instrumentally. The manual container inspection is very time and resource consuming causing delays in shipping and extra costs. There is a great need to inspect shipping containers more effectively, especially after

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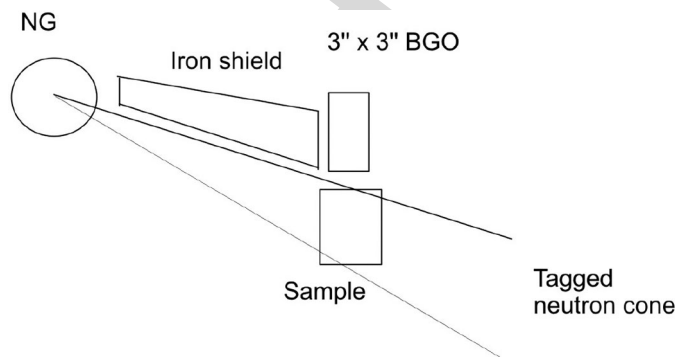


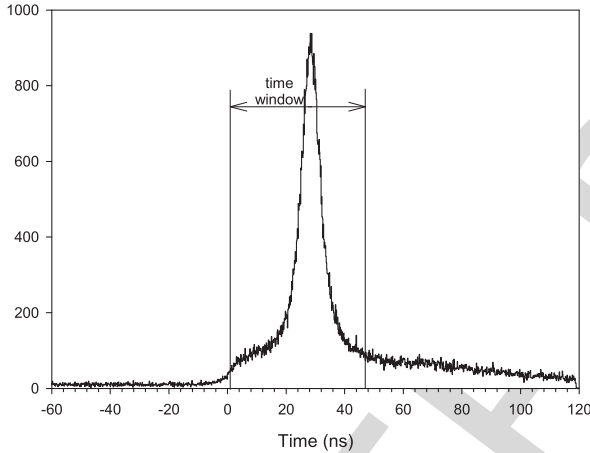
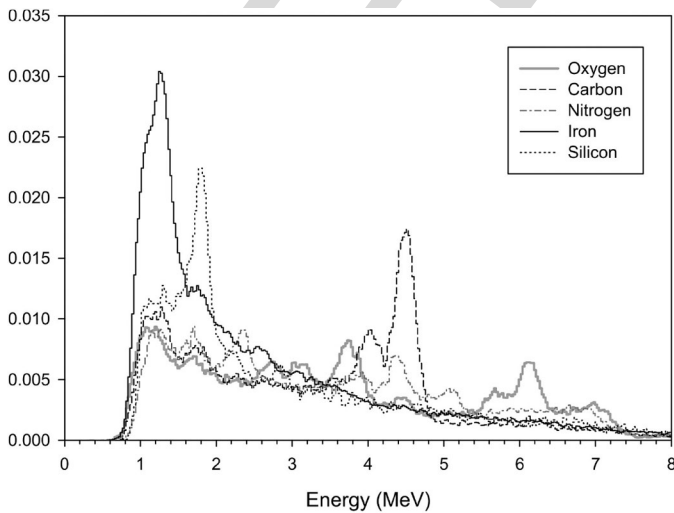
Fig. 1. Sketch of the experimental setup.

F1:1

establishing US 100% inspection regulations [1]. This triggered a lot of research towards development of advanced instrumental inspection methods [2], [3]. The fast neutron activation analysis is one of the most promising non-intrusive elemental analysis techniques for inspection of large volume cargo. It is well known that fast neutrons generated by a neutron source penetrate very deeply in the measured object and collide with the nuclei. Following the inelastic scatter with the fast neutron the nucleus emits characteristic gamma rays which can be measured and analysed. By measuring the carbon-to-oxygen and nitrogen-to-carbon content of the measured target it is possible to identify dangerous and contraband substances [3]. Up to day the methods using fast neutrons are developed to the point that some pilot installations were built like in [3], and there are few commercial products already available at the market [4]. Yet, there are no established QA/QC procedures to control the neutron interrogation systems for preventing false positive or worse, false negative signals that would lead to underestimation of a potential danger. First steps in applying QA/QC procedures are calibration of the neutron inspection system, setting of standard reference materials and then getting the proof of compliance. In this research we were focused on detecting threat material such as bulk and liquid explosives, chemical warfare agents and drugs as a part of the QA/QC procedures. For this purpose simulants of dangerous substances with different C/O and N/C ratios were created. Here we present the calibration procedure done for unshielded material. Since the C/O and N/C values depend on shielding material too, the same procedure should be repeated for different types of shielding (mineral, metal or organic matrix) which requires some a priori knowledge of the surrounding matrix.

T1:1
T1:2TABLE I
COMPONENTS AND CHEMICAL FORMULAE OF THREAT MATERIALS AND THEIR SIMULANTS

Threat materials	Description	Chemical formulae	Chemical formulae of simulant	Simulant mass composition
Hexogen (RDX)	Explosive	$C_3H_6N_6O_6$	$Si_3C_3H_6N_6O_6$	Melamine 0.412, Quartz sand 0.588
TNT	Explosive	$C_7H_5N_3O_6$	$C_7H_6N_3O_6$	Graphite 0.158, Oxalic acid deh. 0.276, Cyanuric acid 0.566
TATP	Explosive	$C_9H_{18}O_6$	$C_9H_{16.9}O_6$	Sucrose 0.844, Paraffin 0.156
DLM2 [7]	Mine simulant	$C_7H_5N_3O_{3.5}$	-	-
Cocaine	Drug	$C_{17}H_{21}NO_4$	$C_{17}H_{21}NO_4$	Melamine 0.069, Graphite 0.243, Sucrose 0.41, Paraffin 0.277
Heroin	Drug	$C_{21}H_{23}NO_5$	$C_{21}H_{23}NO_5$	Melamine 0.057, Graphite 0.294, Sucrose 0.421, Paraffin 0.227
Yperite	Chemical warfare	$C_4H_8Cl_2S$	$C_4H_{8.2-8.4}Cl_2SNa_2$	Paraffin 0.274, Sodium-chloride 0.57, Sulfur 0.156
Tetranitromethane	Liquid explosive	$C(NO_2)_4$	$Fe_{4.9}C_{4/3}(NO_2)_4H_{32/3}$	Iron(III) nitrate nonahydrate 0.762, Melamine 0.238
Peroxide methylethylketone	Liquid explosive	$C_8H_{18}O_6$	$C_8H_{12}O_6$	Sucrose 0.914, Graphite 0.086
Nitromethane	Liquid explosive	CH_3NO_2	$Fe_{1/3}CH_{5/2}NO_2$	Oxalic acid dehydrated 0.398, Melamine 0.266, Iron(III) oxide 0.336
Ethyleneglycol dinitrate	Liquid explosive	$(CH_2ONO_2)_2$	$FeCH_{5/2}NO_3$	Oxalic acid dehydrated 0.238, Melamine 0.159, Iron(III) oxide 0.603

F2:1 Fig. 2. Typical time-of-flight spectrum (output from TAC) with indicated time
F2:2 window used in data analysis.

F3:1 Fig. 3. Gamma spectra of elements used for fitting procedure.

TABLE II
MASS COMPOSITION OF STANDARDS PREPARED FOR C/O CALIBRATION T2:1
T2:2

C/O	Composition
1/3	Graphite 0.118, quartz sand 0.882
1/2	Graphite 0.167, quartz sand 0.833
1	Graphite 0.286, quartz sand 0.714
2	Graphite 0.444, quartz sand 0.556
4	Graphite 0.615, quartz sand 0.385

TABLE III
MASS COMPOSITION OF STANDARDS PREPARED FOR N/C CALIBRATION T3:1
T3:2

N/C	Composition
1/21	Iron(III) nitrate nonahydrate 0.348, graphite 0.652
3/7	Iron(III) nitrate nonahydrate 0.828, graphite 0.172
2.5	Iron(III) nitrate nonahydrate 0.616, melamine 0.384
3.5	Iron(III) nitrate nonahydrate 0.828, melamine 0.172

II. METHODS

Fig. 1 shows the experimental setup. A neutron generator (NG) ThermoElectron API 120 was used as a source of 14 MeV (fast) neutrons (cca 10^7 n/s in 4π) produced by $^3H(d,n)^4He$ nuclear reaction. Since 4He (alpha) particle is produced in opposite direction from the neutron, it is possible to tag and electronically collimate neutrons by detecting coincides between alpha particles and gamma rays emitted from the nuclei in the measured object excited by the inelastic scattering with tagged neutrons [6]. The alpha detector incorporated inside the NG was made from the YAP:Ce scintillator fixed to the NG, and removable photomultiplier tube (PMT). The collimator in front of the PMT defined the tagged neutron cone opening angle of 12° . The $7.62\text{ cm} \times 7.62\text{ cm}$ Bismuth Germanium Oxide, $Bi_4Ge_3O_{12}$ detector (BGO) was used for the detection of characteristic gamma rays. The BGO energy resolution was 7% at 4.44 MeV carbon peak. A pyramid made of iron, 40 cm in length, was used as a shield for detector protection from direct neutron radiation. A measured sample was put below the gamma ray detector 5 cm apart from the detector. The distance between the neutron source and the gamma

T4:1
T4:2TABLE IV
UNFOLDING RESULTS FOR C/O CALIBRATION STANDARDS

C/O	Oxygen (a)	Carbon (b)	Nitrogen (c)	Iron (d)	Silicon (e)	b/a
1/3	0.45 ± 0.007	0.135 ± 0.005	-0.03 ± 0.01	-0.10 ± 0.01	0.54 ± 0.01	0.30 ± 0.01
1/2	0.395 ± 0.008	0.160 ± 0.005	-0.01 ± 0.01	-0.015 ± 0.012	0.47 ± 0.01	0.405 ± 0.015
1	0.326 ± 0.009	0.277 ± 0.006	-0.008 ± 0.01	0.032 ± 0.014	0.37 ± 0.01	0.85 ± 0.03
2	0.259 ± 0.008	0.401 ± 0.006	0.03 ± 0.01	-0.01 ± 0.01	0.31 ± 0.01	1.55 ± 0.05
4	0.181 ± 0.009	0.588 ± 0.007	0.005 ± 0.01	0.02 ± 0.01	0.20 ± 0.01	3.25 ± 0.2

T5:1
T5:2TABLE V
ACTUAL RELATIVE MASS CONCENTRATION OF OXYGEN, CARBON, NITROGEN, IRON AND SILICON FOR C/O CALIBRATION STANDARDS

C/O	Oxygen	Carbon	Nitrogen	Iron	Silicon	Carbon/Oxygen
1/3	0.47	0.12	0	0	0.41	0.25
1/2	0.44	0.17	0	0	0.39	0.39
1	0.38	0.29	0	0	0.33	0.76
2	0.30	0.44	0	0	0.26	1.47
4	0.21	0.61	0	0	0.18	2.90

T6:1
T6:2TABLE VI
UNFOLDING RESULTS FOR N/C CALIBRATION STANDARDS

N/C	Oxygen (a)	Carbon (b)	Nitrogen (c)	Iron (d)	Silicon (e)	c/b
1/21	0.245 ± 0.009	0.622 ± 0.008	0.06 ± 0.01	0.12 ± 0.01	-0.055 ± 0.01	0.096 ± 0.023
3/7	0.676 ± 0.009	0.172 ± 0.006	0.10 ± 0.01	0.075 ± 0.01	0.0131 ± 0.0095	0.59 ± 0.08
2.5	0.480 ± 0.010	0.133 ± 0.006	0.285 ± 0.015	0.11 ± 0.01	0.025 ± 0.01	2.14 ± 0.15
3.5	0.585 ± 0.010	0.068 ± 0.007	0.19 ± 0.02	0.15 ± 0.02	0.02 ± 0.01	3.0 ± 0.5

T7:1
T7:2TABLE VII
ACTUAL RELATIVE MASS CONCENTRATION OF OXYGEN, CARBON, NITROGEN, IRON AND SILICON FOR N/C CALIBRATION STANDARDS

N/C	Oxygen	Carbon	Nitrogen	Iron	Silicon	Nitrogen/Carbon
1/21	0.248	0.652	0.036	0.048	0	0.055
3/7	0.590	0.172	0.086	0.114	0	0.5
2.5	0.439	0.11	0.320	0.085	0	2.9
3.5	0.590	0.049	0.201	0.114	0	4.1

T8:1
T8:2TABLE VIII
UNFOLDING RESULTS FOR MEASURED SIMULANTS OF THREAT MATERIALS

Item (simulants)	Oxygen (a)	Carbon (b)	Nitrogen (c)	Iron (d)	Silicon (e)
Hexogen (RDX)	0.297±0.009	0.126±0.006	0.220±0.01	0.011±0.014	0.35±0.01
TNT	0.45±0.01	0.369±0.007	0.20±0.01	0.007±0.01	-0.002±0.01
TATP	0.45±0.01	0.535±0.008	0.02±0.01	0.02±0.01	-0.04±0.01
DLM2	0.34±0.01	0.35±0.008	0.15±0.02	0.165±0.017	0.01±0.01
Cocaine	0.205±0.009	0.748±0.008	0.082±0.014	0.038±0.015	-0.05±0.01
Heroin	0.24±0.009	0.762±0.008	0.03±0.014	0.033±0.014	-0.04±0.01
Yperite	-	-	-	-	-
Tetranitromethane	0.53±0.01	0.087±0.007	0.222±0.015	0.163±0.015	0.015±0.012
Peroxide methylethylketone	0.488±0.009	0.525±0.007	0.007±0.013	0.017±0.013	-0.02±0.01
Nitromethane	0.447±0.009	0.16±0.006	0.19±0.01	0.21±0.01	0.01±0.01
Ethylene glycol dinitrate	0.408±0.009	0.111±0.006	0.12±0.01	0.37±0.01	0.015±0.01

98 ray detector was 50 cm. All samples had mass of 1 kg, except
99 DLM2 anti-personal mine simulant [7] which mass was 192.6 g
100 and volume $\Phi 80 \text{ mm} \times 34.5 \text{ mm}$.

101 Measured samples, chemical formulae of the real materials
102 and measured simulants (when applicable), as well as
103 compounds used to make simulants are presented in Table I.

104 Each measurement lasted 1000 s and was normalized to
105 the same number of emitted tagged neutrons. The fast output
106 from the alpha detector was fed through the constant fraction
107 discriminator (CFD) and delay line to the STOP of the time-
108 to-amplitude converter (TAC). The fast output from the gamma
109 ray detector was fed through the timing filter amplifier and CFD

to the START of the TAC. A slow signal from the gamma ray
110 detector was fed through the amplifier to the analog-to-digital
111 converter (ADC) incorporated inside the personal computer
112 together with the output from TAC (time spectrum). The ADC
113 was triggered by a single channel analyzer (SCA) output from
114 TAC. Fig. 2 shows the typical time spectrum. A wider time win-
115 dow was used for the gamma ray spectra analysis in order to
116 obtain more statistics (counts) and accordingly smaller error
117 bars in the gamma ray peaks. This improved the detection
118 probabilities.
119

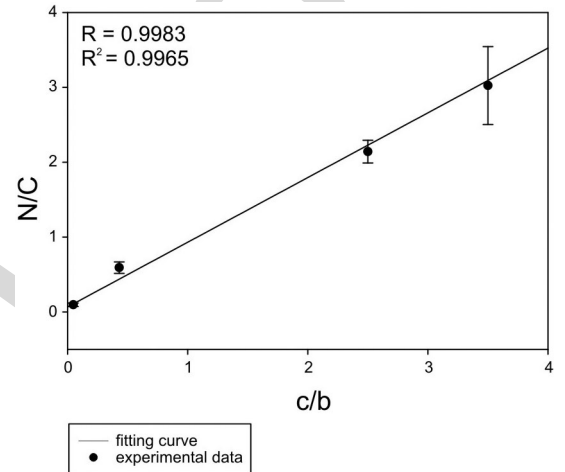
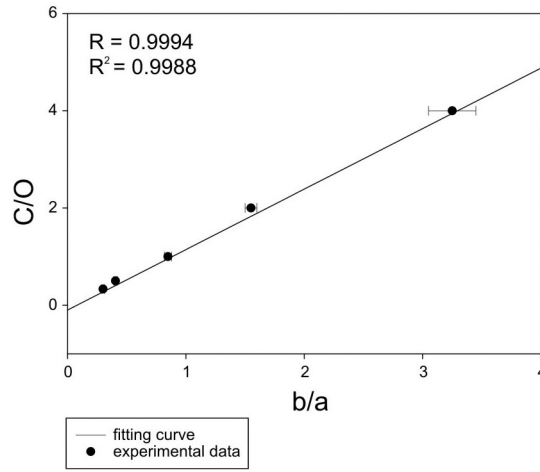
Gamma ray spectra were fitted according to the equation (1)
120 which is simple unfolding model developed for this purpose.
121

T9:1
T9:2

TABLE IX

ACTUAL RELATIVE MASS CONCENTRATION OF OXYGEN, CARBON, NITROGEN, IRON AND SILICON FOR SIMULANTS OF THREAT MATERIALS

Item (simulants)	Oxygen	Carbon	Nitrogen	Iron	Silicon
Hexogen (RDX)	0.313	0.118	0.274	0	0.275
TNT	0.421	0.368	0.184	0	0
TATP	0.434	0.489	0	0	0
DLM2	0.401	0.384	0.192	0	0
Cocaine	0.211	0.673	0.046	0	0
Heroin	0.216	0.683	0.038	0	0
Yperite	-	-	-	-	-
Tetranitromethane	0.543	0.068	0.238	0.105	0
Peroxide methylethylketone	0.470	0.471	0	0	0
Nitromethane	0.404	0.152	0.177	0.235	0
Ethyleneglycol dinitrate	0.362	0.091	0.106	0.422	0

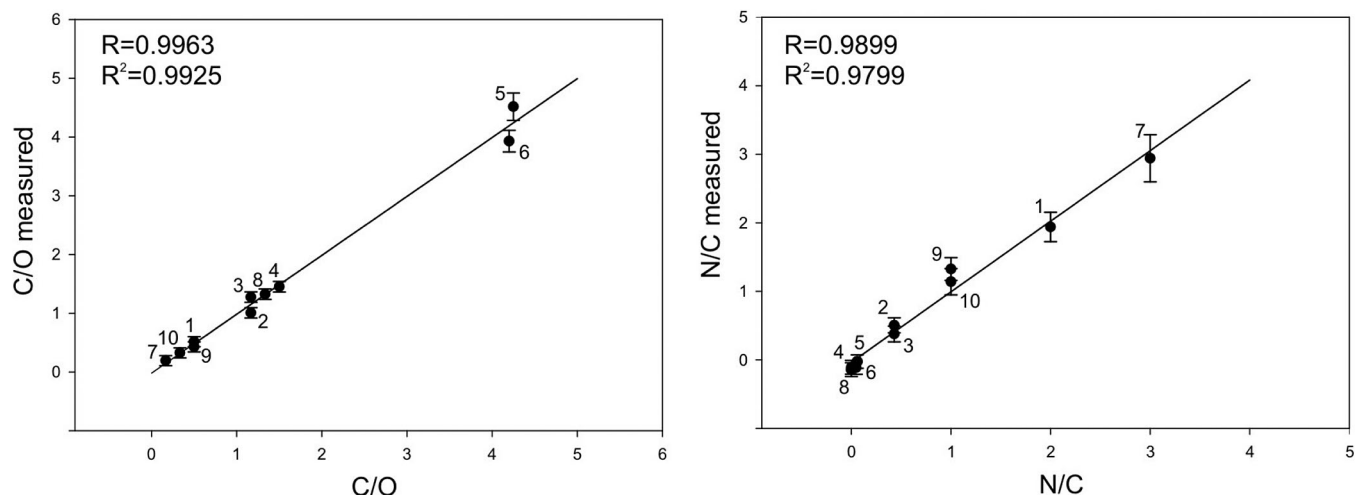


F4:1 Fig. 4. The stoichiometric C/O values in dependence on the fitting parameters
F4:2 ratio for C/O calibration standards. Standard error of estimate: 6%.

Fig. 5. The stoichiometric N/C values in dependence on the fitting parameters
ratio for N/C calibration standards. Standard error of estimate: 12%.

F5:1
F5:2

$$\begin{aligned}
 \chi^2 = & \frac{summ^2}{ch_{\max} - ch_{\min} - 5 + 1} \\
 & \times \sum_{ch=ch_{\min}}^{ch_{\max}} \frac{\left(a \times O(ch) + b \times C(ch) + c \times N(ch) + d \times Fe(ch) + e \times Si(ch) - \frac{T \arg et(ch)}{summ} \right)^2}{T \arg et(ch)} \\
 & \sum_{ch=ch_{\min}}^{ch_{\max}} T \arg et(ch) = summ \\
 & \sum_{ch=ch_{\min}}^{ch_{\max}} O(ch) = 1 \\
 & \sum_{ch=ch_{\min}}^{ch_{\max}} C(ch) = 1 \\
 & \sum_{ch=ch_{\min}}^{ch_{\max}} N(ch) = 1 \\
 & \sum_{ch=ch_{\min}}^{ch_{\max}} Fe(ch) = 1 \\
 & \sum_{ch=ch_{\min}}^{ch_{\max}} Si(ch) = 1
 \end{aligned} \tag{1}$$



F6:1 Fig. 6. Correlations between stoichiometric and experimentally obtained values for C/O ratio (left) and N/C ratio (right) for different types of threat materials as
F6:2 shown in Tables X and XI.

T10:1 TABLE X
T10:2 COMPARISON BETWEEN MEASURED AND STOICHIOMETRIC C/O VALUES
T10:3 OF THREAT MATERIAL SIMULANTS

ID	Simulant	C/O from stoichiometry	C/O from measurement	Difference
1.	Hexogen	0.500	0.514±0.085	-0.16 σ
2.	TNT	1.17	1.01±0.09	1.8 σ
3.	DLM2	1.17	1.27±0.09	-1.1 σ
4.	TATP	1.50	1.45±0.09	0.55 σ
5.	Cocaine	4.25	4.52±0.23	-1.2 σ
6.	Heroin	4.2	3.9±0.2	1.5 σ
7.	Tetranitro-methane	0.167	0.192±0.085	0.3 σ
8.	Peroxide methyl-ethylketone	1.33	1.32±0.09	0.11 σ
9.	Nitromethane	0.500	0.428±0.085	0.85 σ
10.	Ethylenglycol dinitrate	0.333	0.324±0.085	0.1 σ

ID	Simulant	N/C from stoichiometry	N/C from measurement	Difference
1.	Hexogen	2	1.9±0.2	0.5 σ
2.	TNT	0.43	0.5±0.1	-0.7 σ
3.	DLM2	0.43	0.4±0.1	0.3 σ
4.	TATP	0	-0.1±0.1	1 σ
5.	Cocaine	0.059	-0.026±0.098	0.9 σ
6.	Heroin	0.0476	-0.112±0.098	1.6 σ
7.	Tetranitro-methane	3	2.9±0.3	0.3 σ
8.	Peroxide methyl-ethylketone	0	-0.1±0.1	1 σ
9.	Nitromethane	1	1.3±0.2	1.5 σ
10.	Ethylenglycol dinitrate	1	1.1±0.2	0.5 σ

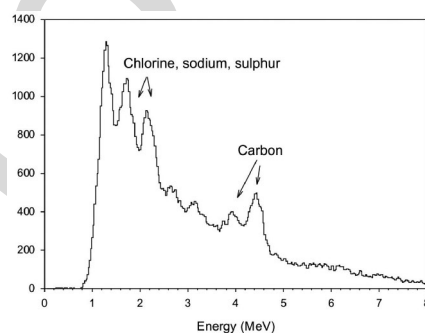


Fig. 7. Gamma spectrum obtained for yperite simulant.

F7:1

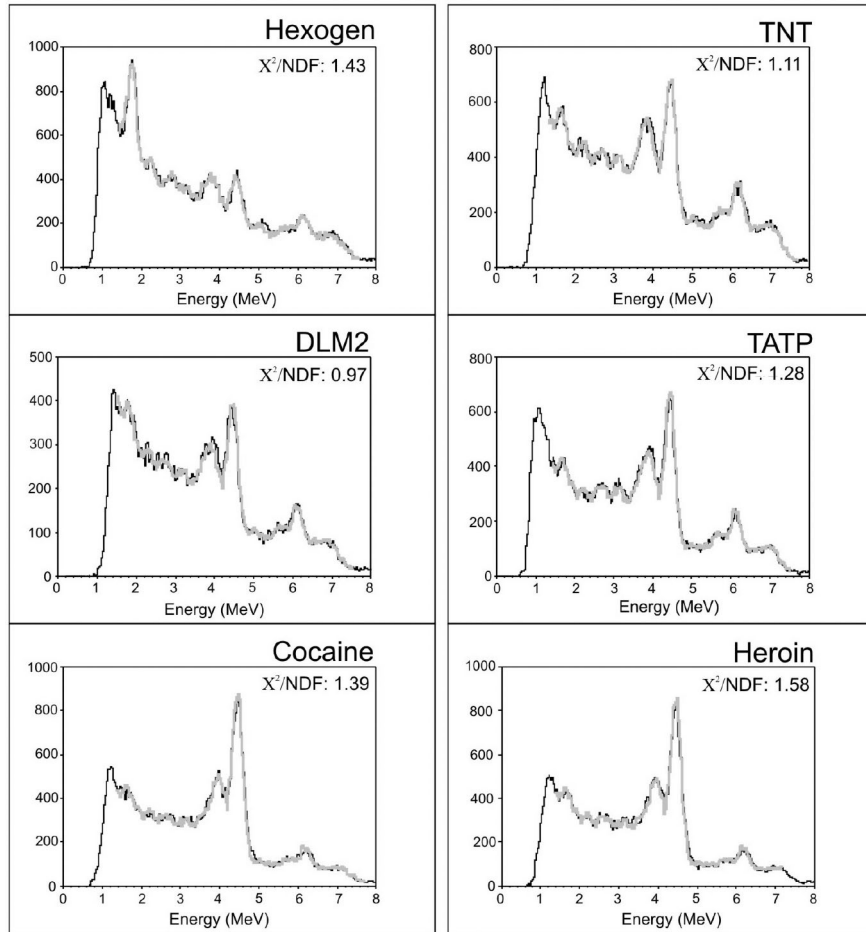
are the one for which the reduced chi-square (defined in eq.1 [8]) is minimal. In practice, values of reduced chi-square close to 1 are acceptable for obtaining a good spectrum fitting. [See equation (1), shown at the bottom of the previous page.]

A number of counts in the gamma ray spectra of carbon are marked with C (ch) and similarly for other elements. A number of counts in the sample spectrum are marked with Target (ch). Spectra obtained for elements oxygen, carbon, nitrogen, iron and silicon are presented in Fig. 3. Oxygen spectrum was obtained by analysis of water, carbon spectrum by analysis of graphite, iron spectrum by analysis of the iron metal plate, nitrogen spectrum by analysis of melamine from which the carbon contribution was subtracted and silicon spectrum was obtained by analysis of quartz sand. In order to quantitatively determine C/O and N/C ratios, standard materials of known C/O and N/C ratios were prepared for calibration purposes as shown in Tables II and III. The C/O and N/C ratios were selected because the differences in their values distinguish the wide spectrum of threat materials.

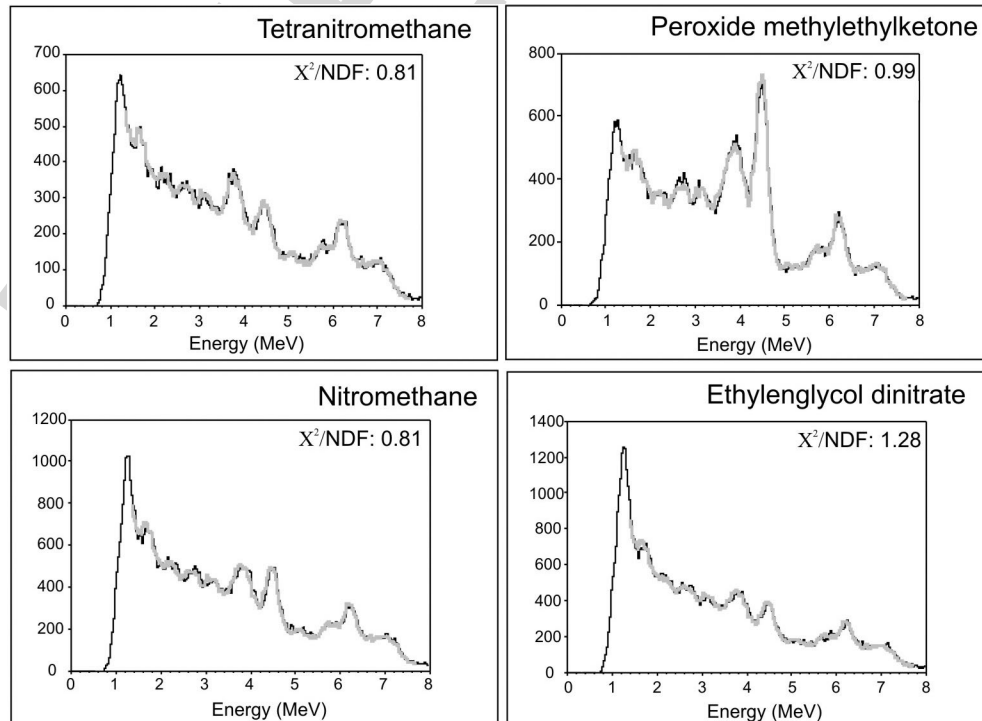
III. RESULTS AND DISCUSSION

Aim of the developed calibration procedure was to relate the measured b/a and c/b ratios with the stoichiometric values of the calibration standards which compositions are shown in Table II

The model assumes that gamma ray spectra contain only the following chemical elements: oxygen, carbon, nitrogen, iron and silicon. The fitting parameters from eq. (1) are marked with “a” for oxygen, “b” for carbon, “c” for nitrogen, “d” for iron and “e” for silicon and the values are summed from minimum to maximum channel and normalized to 1. The method of least squares states that the best value of “a”, “b”, “c”, “d” and “e”



F8:1 Fig. 8. Gamma spectra for simulants of bulk explosives, cocaine and heroin. Fitted spectra are in grey.



F9:1 Fig. 9. Gamma spectra for simulants of liquid explosives. Fitted spectra are in grey.

and III. The method of least squares was used to obtain “a”, “b”, “c”, “d” and “e” fitting parameters with minimum chi-square value which results are presented in Table IV and VI. Negative values of fitting parameters have to be interpreted as zero values within the standard errors of estimate. Actual relative mass concentration of oxygen, carbon, nitrogen, iron and silicon for C/O and N/C calibration standards are given in table V and VII, respectively. Figs. 4 and 5 show the stoichiometric C/O and N/C values in dependence on the fitting parameter ratios b/a and c/b, respectively. Lines were fitted through the experimentally obtained points with the fitting parameters shown in eq. (2) and (3). These calibration lines were used to obtain experimental C/O and N/C values for measured simulants. Fitting parameters obtained for “a”, “b”, “c”, “d” and “e” in simulants are presented in Table VIII and actual relative mass concentrations of O, Si, N, Fe and Si in simulants in Table IX.

Linear correlations between theoretical and experimental ratios of C/O and N/C for hexogen (RDX), TNT, DLM2, TATP, cocaine, heroin, tetranitromethane, peroxide methylethylketone, nitromethane and ethylenglycol dinitrate are shown in eq. (4) and (5) and Fig. 6. Expected slope and segment of the correlation function are 1 and 0, respectively. The total error comprises errors obtained for segment and slope. Since both C/O and N/C ratios have slopes and segments close to 1 and 0 respectively (within the error bar), it can be concluded that the calibration procedure is consistent. Tables X and XI show the comparison between measured and stoichiometric C/O and N/C values respectively, as well as the individual errors resulting from the spectrum fitting. The error bars depend on many factors, however they diminish with the extension of the measurement time. This calibration procedure enables experimental determination of C/O and N/C values by analyzing the adequate standards (Tables II and III) and testing the quality of calibration by analyzing simulants prepared as reference materials (Table I). This approach is different compared to the methodology described in [5] where Monte Carlo simulation was used to determine C/O and N/C values. Applied calibration procedure should be repeated for a shielded material, e.g. for inspected material shielded within metal, mineral or organic matrix having realistic densities obtained from a real cargo [9]. By using this procedure it is possible to obtain correction factors for different types of cargo.

$$C/O(\text{experimental}) = (-0.1 \pm 0.04) + (1.244 \pm 0.025)b/a \quad (2)$$

$$N/C(\text{experimental}) = (-0.16 \pm 0.095) + (1.215 \pm 0.051)c/b \quad (3)$$

$$C/O = (-0.02 \pm 0.07) + (1.0 \pm 0.03)C/O(\text{experimental}) \quad (4)$$

$$N/C = (-0.04 \pm 0.07) + (1.03 \pm 0.05)N/C(\text{experimental}) \quad (5)$$

Fig. 7 presents gamma spectrum obtained for yperite. This spectrum was not fitted since it does not contain N and O peaks. Fig. 8 presents fitted gamma spectra of measured bulk explosives and drugs and Fig. 9 shows fitted spectra of measured liquid explosives. Note that the reduced chi-squares of fitted spectra showed in Fig. 8 and 9 were all close to one, hence the fitting results are acceptable.

IV. CONCLUSION

We have proposed new experimental approach for the neutron interrogation system calibration. Calibration standards and simulants of threat materials were created. Materials and quantities necessary for their production have been listed, thus developers of neutron interrogation systems can produce their own calibration standards and threat material simulants as reference materials. A simple unfolding model for determination of C/O and N/C ratios has been developed. It has been shown that analyses of the gamma ray spectra by using this unfolding model gave a nice agreement with the C/O and N/C stoichiometric ratios of simulants, hence the calibration quality was successfully tested. In future work influence of the surrounding matrix to C/O and N/C ratios in simulants of threat material will be studied.

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Development of the Quality Assurance/Quality Control Procedures for a Neutron Interrogation System

Jasmina Obhodaš, Davorin Sudac, and Vladivoj Valković

Abstract—In order to perform Quality Assurance/Quality Control (QA/QC) procedures for a system dedicated to the neutron interrogation of objects for the presence of threat materials one needs to perform measurements of reference materials (RM) i.e. simulants having the same (or similar) atomic ratios as real materials. It is well known that explosives, drugs, and various other benign materials, contain chemical elements such as hydrogen, oxygen, carbon and nitrogen in distinctly different quantities. For example, a high carbon-to-oxygen ratio (C/O) is characteristic of drugs. Explosives can be differentiated by measurement of both (C/O) and nitrogen-to-oxygen (N/O) ratios. The C/N ratio of the chemical warfare agents, coupled with the measurement of elements such as fluorine and phosphorus, clearly differentiate them from the conventional explosives. Here we present the RM preparation, calibration procedure and correlations attained between theoretical values and experimentally obtained results in laboratory conditions for C/O and N/C ratios of prepared hexogen (RDX), TNT, DLM2, TATP, cocaine, heroin, yperite, tetranitromethane, peroxide methylethylketone, nitromethane and ethyleneglycol dinitrate simulants. We have shown that analyses of the gamma ray spectra by using simple unfolding model developed for this purpose gave a nice agreement with the chemical formula of created simulants, thus the calibration quality was successfully tested.

Index Term



I. INTRODUCTION

THE purpose of container inspection is to compare container cargo with its cargo manifest. Any deviation observed requires further investigation. Container inspection operations are strongly affected by the normal port operations. To this end, it is worthwhile saying that current standards and norms help in identifying suspected containers even before their arrival in the port of destination. However, the inspection of a suspected container can be done only when the container becomes available in the port area, e.g. unloaded from the ship and positioned according to the security officers' request at a specific location in the yard area. The suspected container can be inspected either manually or instrumentally. The manual container inspection is very time and resource consuming causing delays in shipping and extra costs. There is a great need to inspect shipping containers more effectively, especially after

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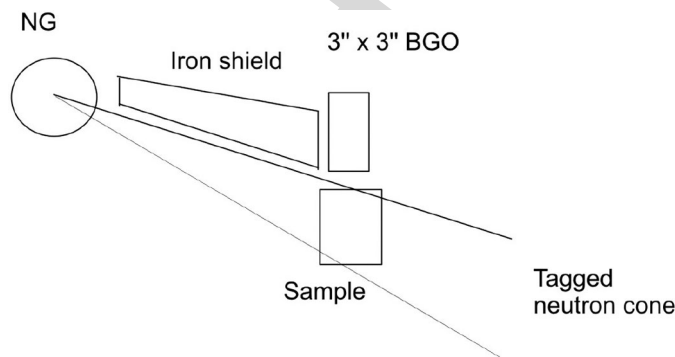


Fig. 1. Sketch of the experimental setup.

F1:1

establishing US 100% inspection regulations [1]. This triggered a lot of research towards development of advanced instrumental inspection methods [2], [3]. The fast neutron activation analysis is one of the most promising non-intrusive elemental analysis techniques for inspection of large volume cargo. It is well known that fast neutrons generated by a neutron source penetrate very deeply in the measured object and collide with the nuclei. Following the inelastic scatter with the fast neutron the nucleus emits characteristic gamma rays which can be measured and analysed. By measuring the carbon-to-oxygen and nitrogen-to-carbon content of the measured target it is possible to identify dangerous and contraband substances [3]. Up to day the methods using fast neutrons are developed to the point that some pilot installations were built like in [3], and there are few commercial products already available at the market [4]. Yet, there are no established QA/QC procedures to control the neutron interrogation systems for preventing false positive or worse, false negative signals that would lead to underestimation of a potential danger. First steps in applying QA/QC procedures are calibration of the neutron inspection system, setting of standard reference materials and then getting the proof of compliance. In this research we were focused on detecting threat material such as bulk and liquid explosives, chemical warfare agents and drugs as a part of the QA/QC procedures. For this purpose simulants of dangerous substances with different C/O and N/C ratios were created. Here we present the calibration procedure done for unshielded material. Since the C/O and N/C values depend on shielding material too, the same procedure should be repeated for different types of shielding (mineral, metal or organic matrix) which requires some a priori knowledge of the surrounding matrix.

T1:1
T1:2

TABLE I
COMPONENTS AND CHEMICAL FORMULAE OF THREAT MATERIALS AND THEIR SIMULANTS

Threat materials	Description	Chemical formulae	Chemical formulae of simulant	Simulant mass composition
Hexogen (RDX)	Explosive	$C_3H_6N_6O_6$	$Si_3C_3H_6N_6O_6$	Melamine 0.412, Quartz sand 0.588
TNT	Explosive	$C_7H_5N_3O_6$	$C_7H_6N_3O_6$	Graphite 0.158, Oxalic acid deh. 0.276, Cyanuric acid 0.566
TATP	Explosive	$C_9H_{18}O_6$	$C_9H_{16.9}O_6$	Sucrose 0.844, Paraffin 0.156
DLM2 [7]	Mine simulant	$C_7H_5N_3O_{3.5}$	-	-
Cocaine	Drug	$C_{17}H_{21}NO_4$	$C_{17}H_{21}NO_4$	Melamine 0.069, Graphite 0.243, Sucrose 0.41, Paraffin 0.277
Heroin	Drug	$C_{21}H_{23}NO_5$	$C_{21}H_{23}NO_5$	Melamine 0.057, Graphite 0.294, Sucrose 0.421, Paraffin 0.227
Yperite	Chemical warfare	$C_4H_8Cl_2S$	$C_4H_{8.2-8.4}Cl_2SNa_2$	Paraffin 0.274, Sodium-chloride 0.57, Sulfur 0.156
Tetranitromethane	Liquid explosive	$C(NO_2)_4$	$Fe_{4.9}C_{4/3}(NO_2)_4H_{32/3}$	Iron(III) nitrate nonahydrate 0.762, Melamine 0.238
Peroxide methylethylketone	Liquid explosive	$C_8H_{18}O_6$	$C_8H_{12}O_6$	Sucrose 0.914, Graphite 0.086
Nitromethane	Liquid explosive	CH_3NO_2	$Fe_{1/3}CH_{5/2}NO_2$	Oxalic acid dehydrated 0.398, Melamine 0.266, Iron(III) oxide 0.336
Ethyleneglycol dinitrate	Liquid explosive	$(CH_2ONO_2)_2$	$FeCH_{5/2}NO_3$	Oxalic acid dehydrated 0.238, Melamine 0.159, Iron(III) oxide 0.603

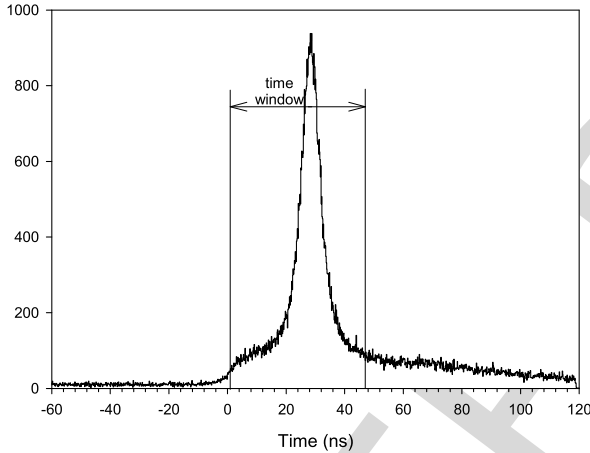


Fig. 2. Typical time-of-flight spectrum (output from TAC) with indicated time window used in data analysis.

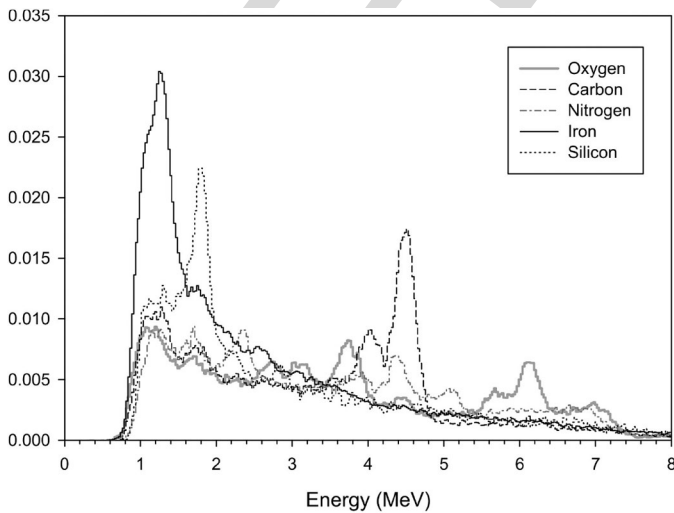


Fig. 3. Gamma spectra of elements used for fitting procedure.

TABLE II
MASS COMPOSITION OF STANDARDS PREPARED FOR C/O CALIBRATION

C/O	Composition
1/3	Graphite 0.118, quartz sand 0.882
1/2	Graphite 0.167, quartz sand 0.833
1	Graphite 0.286, quartz sand 0.714
2	Graphite 0.444, quartz sand 0.556
4	Graphite 0.615, quartz sand 0.385

TABLE III
MASS COMPOSITION OF STANDARDS PREPARED FOR N/C CALIBRATION

N/C	Composition
1/21	Iron(III) nitrate nonahydrate 0.348, graphite 0.652
3/7	Iron(III) nitrate nonahydrate 0.828, graphite 0.172
2.5	Iron(III) nitrate nonahydrate 0.616, melamine 0.384
3.5	Iron(III) nitrate nonahydrate 0.828, melamine 0.172

II. METHODS

Fig. 1 shows the experimental setup. A neutron generator (NG) ThermoElectron API 120 was used as a source of 14 MeV (fast) neutrons (cca 10^7 n/s in 4π) produced by $^3H(d,n)^4He$ nuclear reaction. Since 4He (alpha) particle is produced in opposite direction from the neutron, it is possible to tag and electronically collimate neutrons by detecting coincides between alpha particles and gamma rays emitted from the nuclei in the measured object excited by the inelastic scattering with tagged neutrons [6]. The alpha detector incorporated inside the NG was made from the YAP:Ce scintillator fixed to the NG, and removable photomultiplier tube (PMT). The collimator in front of the PMT defined the tagged neutron cone opening angle of 12° . The $7.62\text{ cm} \times 7.62\text{ cm}$ Bismuth Germanium Oxide, $Bi_4Ge_3O_{12}$ detector (BGO) was used for the detection of characteristic gamma rays. The BGO energy resolution was 7% at 4.44 MeV carbon peak. A pyramid made of iron, 40 cm in length, was used as a shield for detector protection from direct neutron radiation. A measured sample was put below the gamma ray detector 5 cm apart from the detector. The distance between the neutron source and the gamma

T4:1
T4:2TABLE IV
UNFOLDING RESULTS FOR C/O CALIBRATION STANDARDS

C/O	Oxygen (a)	Carbon (b)	Nitrogen (c)	Iron (d)	Silicon (e)	b/a
1/3	0.45 ± 0.007	0.135 ± 0.005	-0.03 ± 0.01	-0.10 ± 0.01	0.54 ± 0.01	0.30 ± 0.01
1/2	0.395 ± 0.008	0.160 ± 0.005	-0.01 ± 0.01	-0.015 ± 0.012	0.47 ± 0.01	0.405 ± 0.015
1	0.326 ± 0.009	0.277 ± 0.006	-0.008 ± 0.01	0.032 ± 0.014	0.37 ± 0.01	0.85 ± 0.03
2	0.259 ± 0.008	0.401 ± 0.006	0.03 ± 0.01	-0.01 ± 0.01	0.31 ± 0.01	1.55 ± 0.05
4	0.181 ± 0.009	0.588 ± 0.007	0.005 ± 0.01	0.02 ± 0.01	0.20 ± 0.01	3.25 ± 0.2

T5:1
T5:2TABLE V
ACTUAL RELATIVE MASS CONCENTRATION OF OXYGEN, CARBON, NITROGEN, IRON AND SILICON FOR C/O CALIBRATION STANDARDS

C/O	Oxygen	Carbon	Nitrogen	Iron	Silicon	Carbon/Oxygen
1/3	0.47	0.12	0	0	0.41	0.25
1/2	0.44	0.17	0	0	0.39	0.39
1	0.38	0.29	0	0	0.33	0.76
2	0.30	0.44	0	0	0.26	1.47
4	0.21	0.61	0	0	0.18	2.90

T6:1
T6:2TABLE VI
UNFOLDING RESULTS FOR N/C CALIBRATION STANDARDS

N/C	Oxygen (a)	Carbon (b)	Nitrogen (c)	Iron (d)	Silicon (e)	c/b
1/21	0.245 ± 0.009	0.622 ± 0.008	0.06 ± 0.01	0.12 ± 0.01	-0.055 ± 0.01	0.096 ± 0.023
3/7	0.676 ± 0.009	0.172 ± 0.006	0.10 ± 0.01	0.075 ± 0.01	0.0131 ± 0.0095	0.59 ± 0.08
2.5	0.480 ± 0.010	0.133 ± 0.006	0.285 ± 0.015	0.11 ± 0.01	0.025 ± 0.01	2.14 ± 0.15
3.5	0.585 ± 0.010	0.068 ± 0.007	0.19 ± 0.02	0.15 ± 0.02	0.02 ± 0.01	3.0 ± 0.5

T7:1
T7:2TABLE VII
ACTUAL RELATIVE MASS CONCENTRATION OF OXYGEN, CARBON, NITROGEN, IRON AND SILICON FOR N/C CALIBRATION STANDARDS

N/C	Oxygen	Carbon	Nitrogen	Iron	Silicon	Nitrogen/Carbon
1/21	0.248	0.652	0.036	0.048	0	0.055
3/7	0.590	0.172	0.086	0.114	0	0.5
2.5	0.439	0.11	0.320	0.085	0	2.9
3.5	0.590	0.049	0.201	0.114	0	4.1

T8:1
T8:2TABLE VIII
UNFOLDING RESULTS FOR MEASURED SIMULANTS OF THREAT MATERIALS

Item (simulants)	Oxygen (a)	Carbon (b)	Nitrogen (c)	Iron (d)	Silicon (e)
Hexogen (RDX)	0.297±0.009	0.126±0.006	0.220±0.01	0.011±0.014	0.35±0.01
TNT	0.45±0.01	0.369±0.007	0.20±0.01	0.007±0.01	-0.002±0.01
TATP	0.45±0.01	0.535±0.008	0.02±0.01	0.02±0.01	-0.04±0.01
DLM2	0.34±0.01	0.35±0.008	0.15±0.02	0.165±0.017	0.01±0.01
Cocaine	0.205±0.009	0.748±0.008	0.082±0.014	0.038±0.015	-0.05±0.01
Heroin	0.24±0.009	0.762±0.008	0.03±0.014	0.033±0.014	-0.04±0.01
Yperite	-	-	-	-	-
Tetranitromethane	0.53±0.01	0.087±0.007	0.222±0.015	0.163±0.015	0.015±0.012
Peroxide methylethylketone	0.488±0.009	0.525±0.007	0.007±0.013	0.017±0.013	-0.02±0.01
Nitromethane	0.447±0.009	0.16±0.006	0.19±0.01	0.21±0.01	0.01±0.01
Ethylene glycol dinitrate	0.408±0.009	0.111±0.006	0.12±0.01	0.37±0.01	0.015±0.01

98 ray detector was 50 cm. All samples had mass of 1 kg, except
99 DLM2 anti-personal mine simulant [7] which mass was 192.6 g
100 and volume $\Phi 80 \text{ mm} \times 34.5 \text{ mm}$.

101 Measured samples, chemical formulae of the real materials
102 and measured simulants (when applicable), as well as
103 compounds used to make simulants are presented in Table I.

104 Each measurement lasted 1000 s and was normalized to
105 the same number of emitted tagged neutrons. The fast output
106 from the alpha detector was fed through the constant fraction
107 discriminator (CFD) and delay line to the STOP of the time-
108 to-amplitude converter (TAC). The fast output from the gamma
109 ray detector was fed through the timing filter amplifier and CFD

to the START of the TAC. A slow signal from the gamma ray
110 detector was fed through the amplifier to the analog-to-digital
111 converter (ADC) incorporated inside the personal computer
112 together with the output from TAC (time spectrum). The ADC
113 was triggered by a single channel analyzer (SCA) output from
114 TAC. Fig. 2 shows the typical time spectrum. A wider time win-
115 dow was used for the gamma ray spectra analysis in order to
116 obtain more statistics (counts) and accordingly smaller error
117 bars in the gamma ray peaks. This improved the detection
118 probabilities.
119

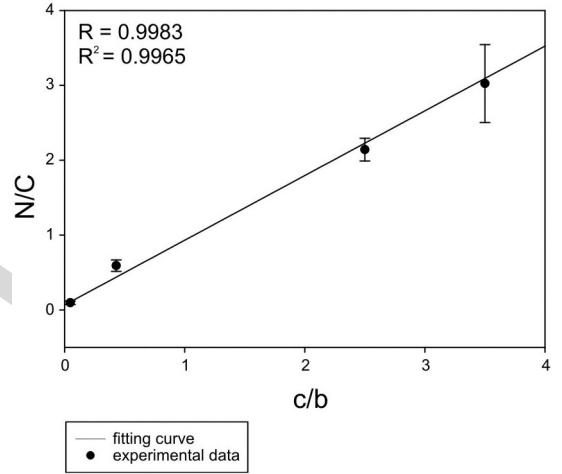
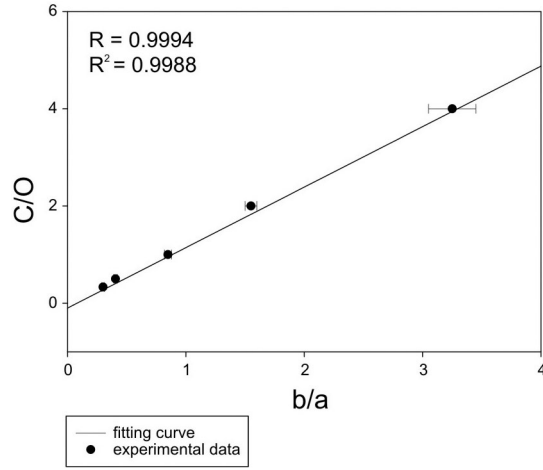
Gamma ray spectra were fitted according to the equation (1)
120 which is simple unfolding model developed for this purpose.
121

T9:1
T9:2

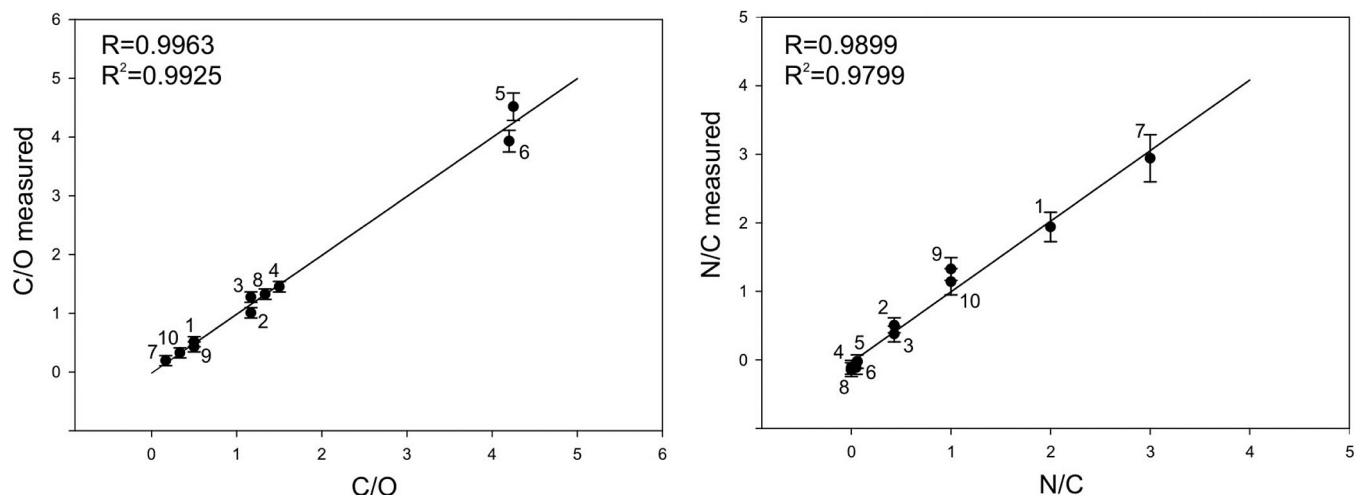
TABLE IX

ACTUAL RELATIVE MASS CONCENTRATION OF OXYGEN, CARBON, NITROGEN, IRON AND SILICON FOR SIMULANTS OF THREAT MATERIALS

Item (simulants)	Oxygen	Carbon	Nitrogen	Iron	Silicon
Hexogen (RDX)	0.313	0.118	0.274	0	0.275
TNT	0.421	0.368	0.184	0	0
TATP	0.434	0.489	0	0	0
DLM2	0.401	0.384	0.192	0	0
Cocaine	0.211	0.673	0.046	0	0
Heroin	0.216	0.683	0.038	0	0
Yperite	-	-	-	-	-
Tetranitromethane	0.543	0.068	0.238	0.105	0
Peroxide methylethylketone	0.470	0.471	0	0	0
Nitromethane	0.404	0.152	0.177	0.235	0
Ethyleneglycol dinitrate	0.362	0.091	0.106	0.422	0

F4:1 Fig. 4. The stoichiometric C/O values in dependence on the fitting parameters
F4:2 ratio for C/O calibration standards. Standard error of estimate: 6%.F5:1 Fig. 5. The stoichiometric N/C values in dependence on the fitting parameters
F5:2 ratio for N/C calibration standards. Standard error of estimate: 12%.

$$\begin{aligned}
 \chi^2 = & \frac{summ^2}{ch_{\max} - ch_{\min} - 5 + 1} \\
 & \times \sum_{ch=ch_{\min}}^{ch_{\max}} \frac{\left(a \times O(ch) + b \times C(ch) + c \times N(ch) + d \times Fe(ch) + e \times Si(ch) - \frac{T \arg et(ch)}{summ} \right)^2}{T \arg et(ch)} \\
 & \sum_{ch=ch_{\min}}^{ch_{\max}} T \arg et(ch) = summ \\
 & \sum_{ch=ch_{\min}}^{ch_{\max}} O(ch) = 1 \\
 & \sum_{ch=ch_{\min}}^{ch_{\max}} C(ch) = 1 \\
 & \sum_{ch=ch_{\min}}^{ch_{\max}} N(ch) = 1 \\
 & \sum_{ch=ch_{\min}}^{ch_{\max}} Fe(ch) = 1 \\
 & \sum_{ch=ch_{\min}}^{ch_{\max}} Si(ch) = 1
 \end{aligned} \tag{1}$$



F6:1 Fig. 6. Correlations between stoichiometric and experimentally obtained values for C/O ratio (left) and N/C ratio (right) for different types of threat materials as
F6:2 shown in Tables X and Y.

T10:1 TABLE X
T10:2 COMPARISON BETWEEN MEASURED AND STOICHIOMETRIC C/O VALUES
T10:3 OF THREAT MATERIAL SIMULANTS

ID	Simulant	C/O from stoichiometry	C/O from measurement	Difference
1.	Hexogen	0.500	0.514±0.085	-0.16 σ
2.	TNT	1.17	1.01±0.09	1.8 σ
3.	DLM2	1.17	1.27±0.09	-1.1 σ
4.	TATP	1.50	1.45±0.09	0.55 σ
5.	Cocaine	4.25	4.52±0.23	-1.2 σ
6.	Heroin	4.2	3.9±0.2	1.5 σ
7.	Tetranitro-methane	0.167	0.192±0.085	0.3 σ
8.	Peroxide methyl-ethylketone	1.33	1.32±0.09	0.11 σ
9.	Nitromethane	0.500	0.428±0.085	0.85 σ
10.	Ethylenglycol dinitrate	0.333	0.324±0.085	0.1 σ

ID	Simulant	N/C from stoichiometry	N/C from measurement	Difference
1.	Hexogen	2	1.9±0.2	0.5 σ
2.	TNT	0.43	0.5±0.1	-0.7 σ
3.	DLM2	0.43	0.4±0.1	0.3 σ
4.	TATP	0	-0.1±0.1	1 σ
5.	Cocaine	0.059	-0.026±0.098	0.9 σ
6.	Heroin	0.0476	-0.112±0.098	1.6 σ
7.	Tetranitro-methane	3	2.9±0.3	0.3 σ
8.	Peroxide methyl-ethylketone	0	-0.1±0.1	1 σ
9.	Nitromethane	1	1.3±0.2	1.5 σ
10.	Ethylenglycol dinitrate	1	1.1±0.2	0.5 σ

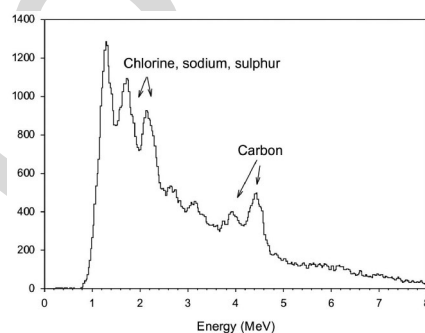


Fig. 7. Gamma spectrum obtained for yperite simulant.

F7:1

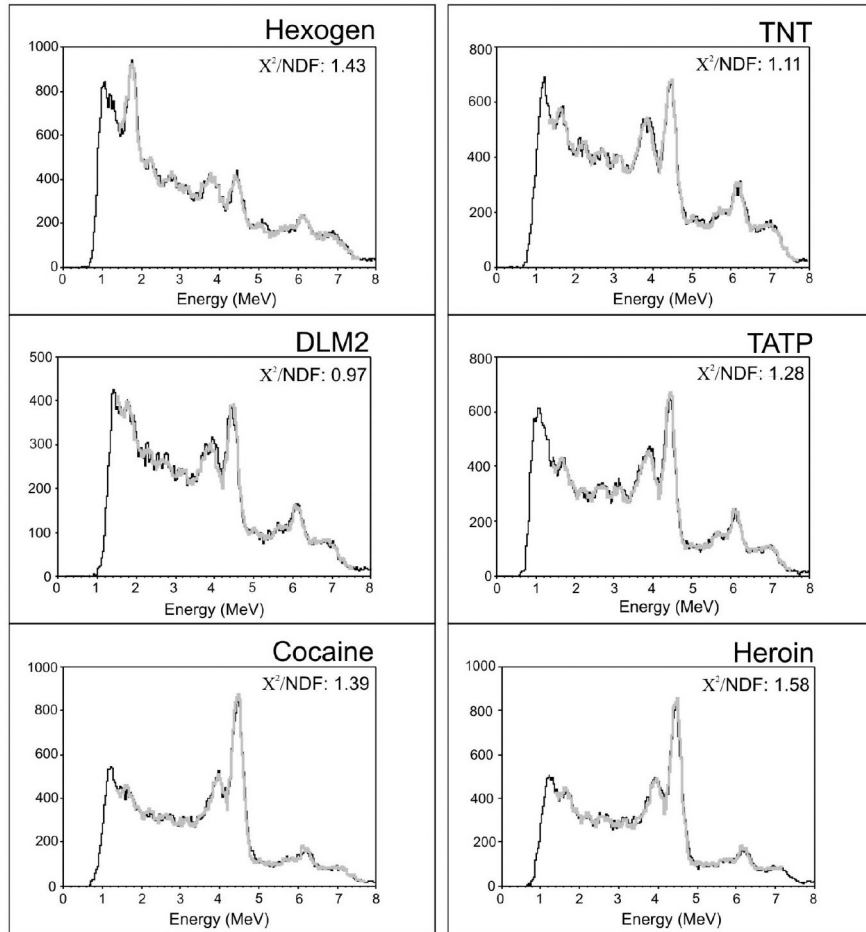
are the one for which the reduced chi-square (defined in eq.1 [8]) is minimal. In practice, values of reduced chi-square close to 1 are acceptable for obtaining a good spectrum fitting. [See equation (1), shown at the bottom of the previous page.]

A number of counts in the gamma ray spectra of carbon are marked with C (ch) and similarly for other elements. A number of counts in the sample spectrum are marked with Target (ch). Spectra obtained for elements oxygen, carbon, nitrogen, iron and silicon are presented in Fig. 3. Oxygen spectrum was obtained by analysis of water, carbon spectrum by analysis of graphite, iron spectrum by analysis of the iron metal plate, nitrogen spectrum by analysis of melamine from which the carbon contribution was subtracted and silicon spectrum was obtained by analysis of quartz sand. In order to quantitatively determine C/O and N/C ratios, standard materials of known C/O and N/C ratios were prepared for calibration purposes as shown in Tables II and III. The C/O and N/C ratios were selected because the differences in their values distinguish the wide spectrum of threat materials.

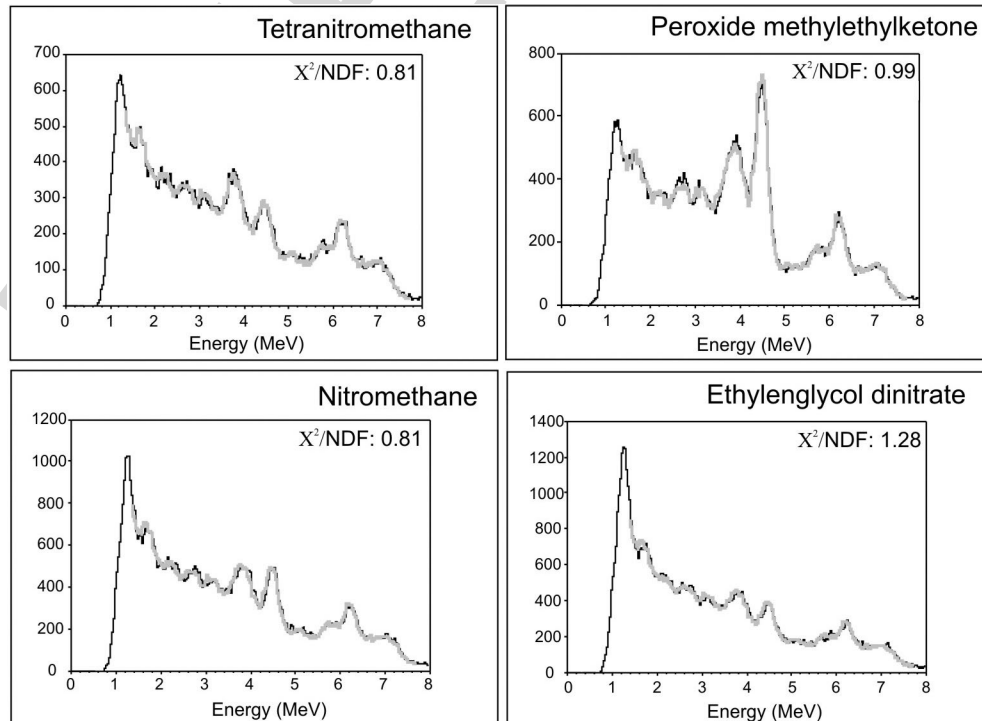
III. RESULTS AND DISCUSSION

Aim of the developed calibration procedure was to relate the measured b/a and c/b ratios with the stoichiometric values of the calibration standards which compositions are shown in Table II

The model assumes that gamma ray spectra contain only the following chemical elements: oxygen, carbon, nitrogen, iron and silicon. The fitting parameters from eq. (1) are marked with “a” for oxygen, “b” for carbon, “c” for nitrogen, “d” for iron and “e” for silicon and the values are summed from minimum to maximum channel and normalized to 1. The method of least squares states that the best value of “a”, “b”, “c”, “d” and “e”



F8:1 Fig. 8. Gamma spectra for simulants of bulk explosives, cocaine and heroin. Fitted spectra are in grey.



F9:1 Fig. 9. Gamma spectra for simulants of liquid explosives. Fitted spectra are in grey.

and III. The method of least squares was used to obtain “a”, “b”, “c”, “d” and “e” fitting parameters with minimum chi-square value which results are presented in Table IV and VI. Negative values of fitting parameters have to be interpreted as zero values within the standard errors of estimate. Actual relative mass concentration of oxygen, carbon, nitrogen, iron and silicon for C/O and N/C calibration standards are given in table V and VII, respectively. Figs. 4 and 5 show the stoichiometric C/O and N/C values in dependence on the fitting parameter ratios b/a and c/b, respectively. Lines were fitted through the experimentally obtained points with the fitting parameters shown in eq. (2) and (3). These calibration lines were used to obtain experimental C/O and N/C values for measured simulants. Fitting parameters obtained for “a”, “b”, “c”, “d” and “e” in simulants are presented in Table VIII and actual relative mass concentrations of O, Si, N, Fe and Si in simulants in Table IX.

Linear correlations between theoretical and experimental ratios of C/O and N/C for hexogen (RDX), TNT, DLM2, TATP, cocaine, heroin, tetranitromethane, peroxide methylethylketone, nitromethane and ethylenglycol dinitrate are shown in eq. (4) and (5) and Fig. 6. Expected slope and segment of the correlation function are 1 and 0, respectively. The total error comprises errors obtained for segment and slope. Since both C/O and N/C ratios have slopes and segments close to 1 and 0 respectively (within the error bar), it can be concluded that the calibration procedure is consistent. Tables X and Y show the comparison between measured and stoichiometric C/O and N/C values respectively, as well as the individual errors resulting from the spectrum fitting. The error bars depend on many factors, however they diminish with the extension of the measurement time. This calibration procedure enables experimental determination of C/O and N/C values by analyzing the adequate standards (Tables II and III) and testing the quality of calibration by analyzing simulants prepared as reference materials (Table I). This approach is different compared to the methodology described in [5] where Monte Carlo simulation was used to determine C/O and N/C values. Applied calibration procedure should be repeated for a shielded material, e.g. for inspected material shielded within metal, mineral or organic matrix having realistic densities obtained from a real cargo [9]. By using this procedure it is possible to obtain correction factors for different types of cargo.

$$C/O(\text{experimental}) = (-0.1 \pm 0.04) + (1.244 \pm 0.025)b/a \quad (2)$$

$$N/C(\text{experimental}) = (-0.16 \pm 0.095) + (1.215 \pm 0.051)c/b \quad (3)$$

$$C/O = (-0.02 \pm 0.07) + (1.0 \pm 0.03)C/O(\text{experimental}) \quad (4)$$

$$N/C = (-0.04 \pm 0.07) + (1.03 \pm 0.05)N/C(\text{experimental}) \quad (5)$$

Fig. 7 presents gamma spectrum obtained for yperite. This spectrum was not fitted since it does not contain N and O peaks. Fig. 8 presents fitted gamma spectra of measured bulk explosives and drugs and Fig. 9 shows fitted spectra of measured liquid explosives. Note that the reduced chi-squares of fitted spectra showed in Fig. 8 and 9 were all close to one, hence the fitting results are acceptable.

IV. CONCLUSION

We have proposed new experimental approach for the neutron interrogation system calibration. Calibration standards and simulants of threat materials were created. Materials and quantities necessary for their production have been listed, thus developers of neutron interrogation systems can produce their own calibration standards and threat material simulants as reference materials. A simple unfolding model for determination of C/O and N/C ratios has been developed. It has been shown that analyses of the gamma ray spectra by using this unfolding model gave a nice agreement with the C/O and N/C stoichiometric ratios of simulants, hence the calibration quality was successfully tested. In future work influence of the surrounding matrix to C/O and N/C ratios in simulants of threat material will be studied.

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