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Charles Crespy, Philippe Duvauchelle, Valerie Kaftandjian, Ferréol Soulez,  
Pascal Ponard

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# Energy Dispersive X Ray Diffraction to identify Explosive Substances : spectra analysis procedure optimization

Crespy C.<sup>1</sup>, Duvauchelle P.<sup>1</sup>, Kaftandjian V.<sup>1</sup>, Soulez F.<sup>1</sup>, Ponard P.<sup>2</sup>

<sup>1</sup>CNDRI – Insa Lyon, Université de Lyon, F-69621, Villeurbanne cedex, France

<sup>2</sup>Thales components and subsystems, 2 rue Marcel Dassault 78491, Velizy cedex, France

*Corresponding author: Charles Crespy*

email: [charles.crespy@insa-lyon.fr](mailto:charles.crespy@insa-lyon.fr); phillipe.duvauchelle@insa-lyon.fr

ad: Insa Lyon, Université de Lyon, CNDRI, F-69621, Villeurbanne cedex, France

tel: 0033472437084

## **Abstract:**

To detect the presence of explosives in packages, automated systems are required. Energy dispersive X-ray diffraction (EDXRD) represents a powerful non invasive tool providing information on the atomic structure of samples. In this paper, EDXRD is investigated as a suitable technique for explosive detection and identification. To this end, a database has been constructed, containing measured X-ray diffraction spectra of several explosives and common materials. In order to quantify spectral resolution influence, this procedure is repeated with two different detectors which have different spectral resolution. Using our database, some standard spectrum analysis procedures generally used for this application have been implemented. Regarding to the results, it's possible to conclude on the robustness and the limits of each analysis procedure. The aim of this work is to define a robust and efficient sequence of EDXRD spectra analysis to discriminate explosive substances. Since our explosive substances are crystalline, the first step consists in using characteristic of the spectrum to estimate a crystallinity criterion which allows to remove a large part of common materials. The second step is a more detailed analysis, it consists in using similarity criterion and major peaks location to differentiate explosive from crystalline common materials. The influence of the spectral resolution on the detection is also examined.

**Key words:** Explosives detection, X-ray diffraction, non destructive testing

## **1. Introduction**

Energy dispersive X-ray diffraction (EDXRD) has been successfully used for many years to provide information about the crystalline structure of samples. In this technique, a sample is probed by a

39 polychromatic X-ray beam. Photon-matter interactions occur along the incident X-ray beam, in  
40 particular, the coherent scattering. The measurement is performed at a fixed scattering angle ( $\theta$ )  
41 with an energy resolved detector. The spectrum thus obtained is material specific since it is linked  
42 to the atomic planar spacing ( $d$ ) and to the radiation wavelength ( $\lambda$ ) according to Bragg law:  $2d\sin$   
43  $\theta = n\lambda$  (where  $n$  is an integer).

44 Nowadays, EDXRD is a well established technique developed for a large number of inspection  
45 applications (see [**Harding'90**] and [**Luggar'99**]). The spectrum analysis procedure is chosen over  
46 the major requirements of the application (fast acquisition, accuracy...). Historically, the procedure  
47 to recognize an unknown substance from its diffraction spectrum has been developed by Hanawalt  
48 in 1936 (see [**Langford'96**]). It is based on comparison between the main peaks location of the  
49 sample spectrum and referenced ones in a paper library.

50 Some particular spectra analysis procedures have been introduced for specific applications. In  
51 [**Luggar'99b**], oil and water ratios are measured in bulk liquid. The analysis spectra procedure  
52 consists in performing ratio of the scattering into two energy windows, which provides results with  
53 a relative error of 0.6%. [**Ballirano'08**] reports the first application of EDXRD to the simultaneous  
54 structural and compositional characterization of papers. The procedure consists in analyzing the  
55 shape of the relevant peak of the crystalline form of cellulose. In [**Farquharson'97**], authors show  
56 that multivariate analysis (MVA) can be a useful technique to estimate components concentration in  
57 bones from EDXRD spectra. Principal components are estimated, then calibration bone phantoms  
58 are constructed to train the MVA model. The results obtained with measured spectra highlight the  
59 influence of exposure time on the accuracy which can reach 3%. For all these studies, the analysis is  
60 based on the assumption that the spectrum under testing contains known materials.

61 In the case of luggages inspection, materials number and type are unknown. They can be divided  
62 into illicit materials (like drugs, explosives) and common ones (such as sugar, toothpaste...). The  
63 identification of illicit materials from their diffraction spectra is usually based on a comparison with  
64 a library. One can distinguish different cases : either the library contains only illicit materials  
65 spectra, or a complete set of illicit and common materials.

66 In [**Cook'07**], X-ray diffraction is proposed as a suitable method to identify illicit drugs in parcels.  
67 To this end, spectra of several drugs and possible cutting agents have been used to train a MVA  
68 software to identify drug in samples. The technique provides quantitative data of a desired variable  
69 (like drug content). In order to reduce the false alarm rate, a database containing the measured  
70 spectra of several thousand materials likely found in parcels has to be used. A specific software has  
71 been developed to predict EDXRD experimental spectra from powder diffraction profiles

72 [Cook'09]. The interest is that powder diffraction files are high energy resolution, and thus, the  
73 software allows to model different experimental conditions (see [Pani'09]). However, the database  
74 must contain a huge number of materials. In [Cook'09b], authors show that MVA predictions based  
75 on simulated data are close to MVA predictions based on measured data.

76 Concerning explosive devices detection inside packages, EDXRD has been examined over the last  
77 decade as a suitable tool. In order to reduce the time measurement and to increase the spectral  
78 resolution, an experimental device geometry has been established to optimize explosive detection  
79 by Luggar and al. (see [Luggar'96] and [Luggar'97]). In [Luggar'98], MVA is performed to  
80 detect the presence of explosives. His study demonstrates how MVA may be applied to reduce  
81 acquisition time which constitutes one of the main requirement for explosive detection. The limit of  
82 this study is the low number of substances used to calibrate the model which leads to a high false  
83 alarm rate. In [Malden'00] EDXRD combined with an angular dispersion is examined. It seems to  
84 be an efficient way to improve robustness but requires a more complex experimental device.  
85 Harding studied the detection of crystalline explosives (see [Harding'99] and [Harding'04]). The  
86 data processing takes place in two stages. At first, Wiener filtering is performed to reduce noise in  
87 spectra recorded with a low acquisition time, then similarity measure between sample spectra and  
88 all library entries is calculated. The author does not disclose the false alarm rate related to this  
89 method. In [Harding'06] and [Harding'07] the author focuses on amorphous explosive detection.  
90 The main conclusions of the Harding's studies are summed up in [Harding'09].

91  
92 The present survey concerning substance identification with EDXRD highlights that different kinds  
93 of analysis procedures could be implemented successfully. In the present study, a database  
94 containing measured EXDRD spectra of several explosives and common materials has been  
95 constructed. To construct the database, two kinds of detector, characterized by their spectral  
96 resolution, are used. In a second time, different classical analysis procedures are implemented, the  
97 robustness and the limit of each methods are examined. Finally we propose a spectra analysis  
98 sequence to discriminate explosive substances. The paper is organized as follow: the first section  
99 deals with the experimental procedure used to build the library, then the recorded spectra are  
100 commented in the second section. Finally the robustness of different analysis procedures are tested  
101 in the third section.

102

## 103 **2.EDXRD technique**

104

### 105 **2.1. Experimental arrangement**

106

## Figure 1

107  
108  
109 Schematic representation of experimental device is shown in **figure 1**. X-ray radiations from a 100  
110 kV tungsten anode X-ray tube operating at 280  $\mu$ A irradiate the sample. The X-ray source has a 10  
111  $\mu$ m focal spot. The scattered photons are detected with detector orientated at  $4.28^\circ$  from the incident  
112 beam. Two types of detectors are used to acquire spectra in this work: *CdZnTe* or High purity  
113 Germanium (*Ge*). The main difference between both detectors is that *Ge* detector has to be cooled  
114 by cryogenic system (it cannot be used in a mobile detection system,) while *CdZnTe* detector is a  
115 room temperature detector. This point leads to a better spectral resolution when *Ge* detector is used.  
116 *CdZnTe* detector has a FWHM (full width half middle) of 2 keV at 60 keV (see [Verger'07]), while  
117 *Ge* detector has a FWHM of 0.4 keV at 60 keV Another difference between spectra acquired with  
118 *CdZnTe* and *Ge* is the difference between spectral range of both detectors. It can be noticed that the  
119 measuring range of *Ge* detector is from 15 to 100 keV while the measuring range of *CdZnTe*  
120 detector is from 30 to 100 keV. The *CdZnTe* detector cut the spectra around 30 keV, this limit is not  
121 due to the crystal but just an electronic threshold. In the case of luggage inspection, this threshold is  
122 out consequence because low energy photons are strongly attenuated thus information under 40 keV  
123 is not really useful.

124 Collimation system allows the direct localization of a scatter volume within the sample. The  
125 collimator located after the source is a slit of 0.2 mm width and 8 cm length. The collimator located  
126 in front of the detector is a slit of 0.1 mm width and 8 cm length. The scatter angle is a critical  
127 parameter which affects the energy resolution, the peaks intensity. It is chosen close to  $4^\circ$  as a good  
128 compromise between angular resolution and main peaks location of explosive substances.  
129 Scattering angle is estimated accurately thanks to graphite spectrum measurement performed with  
130 *Ge* detector. The uncertainty of the scatter angle is  $\pm 0.05^\circ$ . Finally, the exposure time is set at  
131 600s, for each spectra spectra to have a good counting statistic.

132

## 133 2.2 EDXRD Spectra

134

135 In EDXRD, the spectra acquired depends not only on the sample but also on the experimental set  
136 up. An example of an EDXRD acquired with *Ge* detector spectrum is presented in **figure 2**.

137

## Figure 2

138 On **figure 2**, it can be noted that the spectra is composed of a baseline and peaks. The baseline is  
139 caused by the amorphous part of the sample, the incoherent scattered radiation and the spectral  
140 shape of X-ray beam. In our configuration, the number of photons whose energy is upper than 90

141 keV is too low to provide suitable information in this range of energy. Thus the useful range of  
142 energy is from 20 keV to 90 keV. The spectral shape of the X-Ray source bias the intensity of the  
143 diffraction peaks too: its influence can be removed by normalization to the transmitted spectrum if  
144 required.

145 Regarding to the peaks, two kinds of peaks must be distinguished. Some are caused by the  
146 characteristic X-ray lines of the X ray tube tungsten target, others are diffraction peaks. Diffraction  
147 peaks are the part of the information which is used to identify the sample. The diffraction peaks  
148 width are linked to the angular dispersion due to the collimation system and the spectral resolution  
149 of the detector. Assuming that diffraction peaks are Gaussian type it can be written:

$$150 \quad FWHM_T = (FWHM_D^2 + FWHM_C^2)^{0.5} \quad (1)$$

151 Where  $FWHM_T$  is the total diffraction peak FWHM,  $FWHM_C$  is the contribution of the collimation  
152 system and  $FWHM_D$  is the contribution of the detector. While  $FWHM_D$  is known,  $FWHM_C$  can be  
153 estimated experimentally by comparing the width of the diffraction peaks with the width of the  
154 fluorescence peaks or numerically by using a Monte Carlo simulation code.

155 The  $FWHM_T$  is experimentally estimated: using a *CdZnTe* detector the FWHM of a diffraction peak  
156 is close to 2.4 keV, while it's close to 0.95 keV for *Ge* detector. These values are of the same order  
157 of magnitude than the numerical prediction. In our case it can be noticed that when *CdZnTe* detector  
158 is used, the detector resolution is the predominant parameter in overall spectral resolution. In the  
159 case of the *Ge* detector, the angular resolution due to the collimation system, is the predominant  
160 parameter.

161 Concerning the noise it can be reduced by rising the exposure time. In the case of an automated  
162 system the exposure time has to be as smaller as possible. As it has been advocated by Harding, the  
163 Wiener filter is a suitable tool for noise reduction in EDXRD spectra. The influence of the Wiener  
164 filter on *Ge* and *CdZnTe* detectors spectra is shown on **figure 3**.

165 **Figure 3**

166 In the following all spectra presented have been filtered.

167

### 168 **3. Database diffraction spectra**

169

#### 170 **3.1. Explosives spectra**

171

172 The spectra of five explosive substances have been recorded: *TNT* (100% Tolite), *Semtex* (91 %  
173 RDX), *Formex* (87% PETN), *PLA NP* (89% PETN) and *HCG* (98% RDX). The acquisition time is  
174 set to 600 seconds. As an illustration, the filtered spectra of *PLA NP* and *Formex* recorded with *Ge*

175 and *CdZnTe* detectors are shown in **figure 4**.

176 **Figure 4**

177 The spectra obtained with the *Ge* detector show a serie of well resolved peaks. The spectra acquired  
178 with the *CdZnTe* detector show a series of broader peaks which does not allow to resolve very close  
179 peaks. **Figure 4** shows the influence of the *CdZnTe* hardware cut at 40 keV. *Formex* and *PLA NP*  
180 are characterized by the same explosive substance (*PETN*), this results in two peaks in common  
181 around 45 keV which can be seen in *Ge* spectra.

182

### 183 **3.2. Common material spectra**

184

185 In order to evaluate the robustness of the method, spectra of eleven common materials likely to be  
186 found in packages have been recorded: *NaCl*, *graphite*, *PVC*, *toothpaste*, *sugar*, *cotton*, *soap*,  
187 *aluminum*, *nickel*, *copper* and *coffee*. Filtered spectra of *PVC* and *NaCl* recorded with *Ge* and  
188 *CdZnTe* detectors are shown in **figure 5**. Unlike explosive spectra, it can be noticed that most of  
189 common materials from our library do not show well resolved peaks, but an amorphous trend with  
190 broad peaks.

191 **Figure 5**

192

### 193 **3.3. Uncertainty estimation on peaks location**

194

195 Since peaks location of a spectra is a capital information about the sample, the uncertainty on the  
196 major peak location is estimated. Ten spectra of the same sample successively recorded under  
197 similar conditions are compared. The standard deviation on peak location ( $s$ ) is estimated and used  
198 to calculate the uncertainty ( $2s$ ). This procedure is repeated with three samples (*NaCl*, *Graphite* and  
199 *Aluminum*), for the both detectors, with and without Wiener filtering. The results are shown in  
200 **figure 6**.

201 **Figure 6**

202 It can be concluded that Wiener filtering is not only a means to remove noise, but also an efficient  
203 tool to reduce the uncertainty on peaks location since it allows to reduce significantly the  
204 uncertainty with both detectors. After filtering, the uncertainty on peak location for *Ge* detector is  
205 less close to 0.2 keV instead of 1.6 keV for *CdZnTe* detector. Thus, it is expected to obtain a lower  
206 false alarm rate when *Ge* detector is used.

207

## 208 **4. Spectra analysis procedure**

209

#### 210 **4.1. Crystallinity criterion**

211

212 It can be useful to define a crystallinity criterion to distinguish amorphous materials from  
213 crystalline ones. The chosen method is based on the crystallinity degree estimation as performed  
214 with angular dispersive X ray diffraction (see [Cheetham'98]). A smooth curve which connects  
215 peak base is estimated using mathematical morphology opening technique (see **figure 7**).

216

#### **Figure 7**

217 The area above the smooth curve corresponds to the crystalline part while the area under the smooth  
218 curve corresponds to the amorphous part. The crystallinity criterion is then estimated as the ratio of  
219 upper spectrum area to total diffraction spectrum area. This criterion corresponds to the crystallinity  
220 degree used in Angular dispersive X ray diffraction. In the EDXRD case even if the source  
221 spectrum shape plays an important role we decide to use the same criterion. The quantitative results  
222 of our samples are shown on **figure 8**. Choosing a crystallinity criterion threshold around 0.2 allows  
223 to eliminate a large part of common materials (4 common materials have a crystallinity criterion  
224 higher than 0.2) Thus, this crystallinity criterion estimation can be used as a first discriminative  
225 step.

226

#### **Figure 8**

#### 227 **4.2. Similarity criterion**

228

229 As a second step, it has been chosen to identify explosives by calculating a similarity criterion i.e.  
230 the cross correlation coefficient.

##### 231 *4.1.1. Acquisition time influence*

232 In order to estimate the robustness of the technique, the influence of acquisition time is investigated.  
233 For many substances, six spectra of a same material are acquired with different acquisition times  
234 (60s, 300s, 420s, 600s, 720s and 900s). Then, the cross correlation coefficient is calculated between  
235 the spectrum acquired in 900s and the five others. The threshold in terms of cross correlation  
236 coefficient is fixed at 0.97, i.e. if the cross correlation coefficient between two spectra is higher than  
237 0.97, the spectra are considered identical. The results for *Semtex* spectra with and without filtering  
238 are shown on **figure 9**.

239

#### **Figure 9**

240 This example is typical of all investigated cases. The results show that after filtering, spectra  
241 acquired with different acquisition time are equivalent. Thus, Wiener filter is a suitable tool to



242 reduce acquisition time. The results are the same whatever the detector used.

#### 243 4.1.2. Substance identification

244 To test the method robustness, a library containing the filtered spectra of each materials is  
245 constructed. For a given and unknown material, the cross correlation coefficient is calculated and  
246 compared to each library's spectrum in an identification goal. It can be noted that the whole  
247 spectrum (amorphous and crystalline part) is used for cross correlation analysis. The results are  
248 shown in **Table 1**.

#### 249 **Table 1**

250 Using the *Ge* detector allows to identify a material whatever its crystallinity criterion. Using the  
251 *CdZnTe* detector, the low spectral resolution leads to a partial loss of information. Indeed, although  
252 there is no real false alarm case, confusion between some amorphous materials can be noted. For  
253 instance, *toothpaste* can not be distinguished from *PVC*, nor *cotton* from *coffee*. The confusion  
254 between *TNT* and *Formex* is the only confusion between two substances whose cristallinity criterion  
255 is upper than 0.2. Thus, it can be thought that some crystalline common materials could also lead to  
256 false alarm.

257

#### 258 4.3. Multivariate Calibration

259

260 Detailed description of multivariate calibration procedure can be found in [**Cook'07**] or  
261 [**Farquharson'97**]. The MVA consists in creating a model in order to link experimental data (**X**) to  
262 quantitative information concerning the sample (**Y**). As a first step, a linear regression model of the  
263 form  $\mathbf{Y}=\mathbf{B}\mathbf{X}$  is estimated. To this end, a set of calibration spectra is used to train the model. The  
264 second step consists in using the model to identify an unknown spectrum. In our case, the  
265 experimental **X** data are the measured spectra. While, in general, **Y** data corresponds to composition  
266 of the sample (see [**Cook'07**] [**Luggar'98**]). Thus **X** is a *N* dimension vector (*N* corresponds to the  
267 number of channels), **Y** is a *M* dimension vector (*M* corresponds to the number of substances  
268 searched for). In practice, it's impossible to perform a well conditioned regression in these  
269 conditions, thus **X** variables is reduced to a short number of principal components (PCs). Authors  
270 show that MVA is a very powerful tool when the spectra of the substances to identify have been  
271 used to train the model. Thus, for an unknown package, a database containing several thousands  
272 spectra would be required. In our application the presence of explosive is examined, thus **Y** is not  
273 the sample composition but a threat factor whose value varies from 0 to 1 (0: no explosive in the  
274 sample, 1: explosive in the sample). This kind of model is an efficient way to search for a  
275 characteristic in explosive spectra which could accurately discriminate them from common

276 materials spectra. The model is trained with a set of 84 measured spectra (spectra of the 16  
277 materials acquired with the 5 acquisition time), the number of PCs is 9. The threshold for detecting  
278 the presence of an explosive is fixed to 0.5. Results of the model are shown in **figure 10**.

279 **Figure 10**

280 It can be concluded that this model leads to one mistake whatever the detector used. In order to  
281 check the robustness, the model is tested with spectra from materials which have not been used to  
282 train the model. For instance, the model results for water spectra is a false alarm ( $Y(H_2O)=0.8$ ).  
283 These findings allow to conclude that it's difficult to identify explosives spectra with a linear model.  
284 Thereby, this kind of model is not suitable to discriminate explosive spectra from common  
285 materials. The only way to reduce the false alarm rate should be to train the regression model with a  
286 complete library containing every common materials spectra which is impossible.

287

#### 288 **4.4. Major peaks location analysis**

289

290 Another technique inspired from Hanawalt procedure consists in using the major peaks location in  
291 order to identify a substance. To perform that a Levenberg-Marquardt minimization algorithm is  
292 used to find the sum of Gaussian functions which fit best the spectrum (see **figure 11**). It's to note  
293 that the amorphous part of the spectrum is subtracted before performing the extraction peak  
294 procedure.

295 **Figure 11**

296 The standard deviation of the Gaussian function is linked to the detector spectral resolution and the  
297 collimation characteristics (see §2.2). According to Bragg's law, it must be noted that the standard  
298 deviation of the Gaussian function is not constant but increases with energy. In first approximation,  
299 this point is neglected in the peaks extraction procedure. If a peak corresponds to a characteristic  
300 line of the X ray tungsten, it is not taken into account during the analysis.

301 Position is given in momentum transfer ( $x=\sin(\theta/2)/\lambda$ ) which is a convenient way to express results  
302 independently on the diffraction angle .The two main peaks location from *Ge* spectra of each  
303 sample are plotted in **figure 12**.

304 **Figure 12**

305 It can be noted that the spectra of the explosive substances have the main peak located between 1  
306  $\text{nm}^{-1}$  and  $1.4 \text{ nm}^{-1}$  (respectively 30 keV and 50 keV when  $\theta=4.28^\circ$ ). Sugar, soap and graphite also  
307 show this characteristic. Using two main peaks allows to reduce the false alarm rate: materials  
308 which have the two main peaks located between  $1 \text{ nm}^{-1}$  and  $1.6 \text{ nm}^{-1}$  (respectively 30 keV and 55

309 keV when  $\theta=4.28^\circ$ ) are explosives only.

310 Regarding now *CdZnTe* spectra, the explosive main peaks are located between  $1.2 \text{ nm}^{-1}$  and  $1.6 \text{ nm}^{-1}$   
311  $^{-1}$  (respectively 40 keV and 52 keV when  $\theta=4.28^\circ$ ). Some spectra of common materials (soap, sugar,  
312 graphite) have the same characteristics.

313

### Figure 13

314 The example of graphite is an interesting case. It's known that theoretically the graphite spectrum  
315 has just one peak. When a measured peak is broader than the previsions, peaks extraction procedure  
316 can lead to the detection of two very close peaks. On **figure 3** it can be noted that the main peak of  
317 graphite has a non Gaussian shape (asymmetric shape). This asymmetry is related to the height of  
318 the collimator which increases the angular aperture with angles above the nominal angle. Moreover,  
319 since the graphite peak is located at high energy, it can be though that this point is linked to the  
320 variation of standard deviation of Gaussian fit with energy.

321 To sum up, it can be noted that the couple defined by the two main peaks location is material  
322 specific whatever the detector used. Moreover all explosives spectra of our library have their main  
323 peak located between 1 and  $1.6 \text{ nm}^{-1}$ . Concerning *Ge* acquisition, a second peak in this range of  
324 energy can be detected. In the future it 's important to check that this observation is valid for every  
325 crystalline explosive substances. However, it's possible that others common materials can have  
326 these characteristics, probably crystalline organic materials such as drugs.

327

### 328 4.5 Influence of attenuation on explosive detection

329

330 The aim of this section is to examine if the detection procedure presented above is still valid in the  
331 context of our application (when the explosive is in a package). The main problem concerns photon  
332 attenuation which depends on their energies and then can significantly modify the diffraction  
333 spectrum. Thus, this section focuses on the possibility to identify an explosive spectrum which is  
334 modified by a attenuation. The part of the transmitted signal integrated along the wave length can  
335 be measured using a scintillator.

336 An experimental procedure has been implemented. It consists in measuring the spectra of an  
337 explosive without attenuation, then the experience is repeated when the material is in a package. For  
338 example, **Figure 14**, we can see the presence of explosives (*Semtex*) into a package containing  
339 paper, books and cotton.

340

### Figure 14

341 Six packages containing materials more or less absorbent (cotton, pound, copper ...) are used. Each  
342 package is characterized by the percentage of attenuation it causes. The most mitigating package

343 absorbs 89% of the total transmitted signal while the least absorbs 15% of the signal. The **figure 15**  
344 show the influence of attenuation on *Semtex* spectrum when the transmitted signal is close to 65 %.

345 **Figure 15**

346 We observe on **figure 15** the difficulty to recognize attenuated from original spectrum. This is due  
347 to the fact that attenuation is not spectrally homogeneous, the high energy signal is less attenuated  
348 than the signal at low energy. Fortunately, the peaks location is not modified by the attenuation.  
349 However, it had be found experimentally that when the transmitted signal is below 30%, the peaks  
350 located below 50 keV are no longer detectable. Since the characteristic peaks of explosives are  
351 between 30 and 50 keV, it is considered that explosive detection is not possible if the transmitted  
352 signal is less than 30%.

353 To quantify the influence of attenuation on the cross correlation coefficient identification procedure  
354 the cross correlation coefficient is calculated between the spectrum unattenuated and the attenuated  
355 one. The results are presented in **Figure 16**.

356 **Figure 16**

357 Note that when the transmitted signal is below 65%, it's no longer possible to recognize the  
358 spectrum of *Semtex* using the correlation coefficient. Since attenuation can damage the spectra  
359 shape of a sample, the thresholds of all detection procedures have to be redefined as a function of  
360 attenuation.

361

## 362 **5 Spectra analysis sequence**

363

364 Taking into account the conclusions of the section 4, an algorithm dedicated to automatic detection  
365 has been implemented. The principle is illustrated in **figure 17**. The two entries of the algorithm is  
366 sample spectra and the integrated attenuation.

367 **Figure 17**

368 First, the integrated attenuation is used to define the detection reliability. This parameter influence  
369 the detection thresholds of the detection procedure. If the transmitted signal is less than 30% of the  
370 initial signal, it is considered that the identification is not possible.

371 If the detection reliability is sufficient, the sample spectrum is analyzed. The sample spectrum is  
372 first filtered, then the crystallinity criterion is calculated. If the spectrum of the sample is of  
373 crystalline type (crystallinity criterion  $>0.2$ ), the cross correlation coefficient is used to compare the  
374 sample spectra with the explosive spectra of our library. Finally, in case of non detection, peaks  
375 location of the sample are compared with those of explosives taking into account the uncertainty.

376 It's to note that the whole spectrum (amorphous and crystalline part) is used in cross correlation  
377 analysis, while only the crystalline part is used for peak location analysis. In case of using *CdZnTe*  
378 spectra, only the two main peaks are used for identification while when *Ge* detector is used it's  
379 possible to compare a larger number of peaks in order to reduce the false alarm rate. This step is  
380 important to detect explosive substance in inhomogeneous voxel. The three step detection  
381 procedure allow to increase the robustness. Moreover, the graphic user interface (GUI) allows a  
382 direct visual comparison between sample spectra and those of the library. To illustrate, **Figure 18**  
383 shows a screen-shot of the graphic user interface.

384 **Figure 18**

## 385 **6. Conclusion**

386  
387 In this study, explosive detection using EDXRD has been investigated using a library containing  
388 five explosives and eleven common materials. As a first step it was chosen to study crystalline  
389 explosives. Thus a crystallinity criterion calculation is proposed in order to discriminate them from  
390 common materials. This step allow to discriminate easily many common materials from explosive  
391 ones. To perform a more accurate discrimination three classical spectra analysis procedures have  
392 been implemented: similarity criterion calculation, MVA and main peaks location. It has been  
393 shown that the use of MVA requires to build a complete library containing not only explosives  
394 spectra but also every common materials spectra. That's why this technique is considered as not  
395 suitable for package inspection. On another hand, explosive spectra show some discriminant  
396 characteristics in term of peaks location and cross correlation coefficient. Whatever the analysis  
397 procedure, the *Ge* spectra are more suitable for discrimination than *CdZnTe*. Thus *CdZnTe detector*  
398 can be applied as a complementary technique in a portable system in order to reduce the false alarm  
399 rate.

400 Following this study, the library will be completed with other explosive spectra, and the detection  
401 technique will be tested with more crystalline common materials in order to estimate accurately the  
402 false alarm rate. Moreover, to improve the discrimination rate with *CdZnTe* detector it's envisaged  
403 to decrease the scattering angle at  $3^\circ$  and redefine the collimation system.

404

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466 **Figures caption:**

467 **figure 1:** schematic representation of the experimental setup

468 **figure 2:** Example of a diffraction spectrum acquired with *Ge* detector

469 **figure 3:** visualization of Wiener filter influence on measured spectra with *CdZnTe* (a) and with *Ge*  
470 (b)

471 **figure 4:** examples of explosive spectra measured with a *Ge* detector (left) and a *CdZnTe* detector  
472 (right)

473 **figure 5:** examples of common materials spectra measured with a *Ge* detector and a *CdZnTe*  
474 detector

475 **figure 6:** peak location uncertainty

476 **figure 7:** estimation of amorphous area (gray zone) with morphological opening of the global  
477 spectrum (on the left for the PLA NP and on the right for PVC)

478 **figure 8:** cristallinity criterion of our samples

479 **figure 9:** cross correlation coefficient as a function of exposure time for *Semtex* and the 2 studied  
480 detectors

481 **figure 10:** MVA results for *CdZnTe* and *Ge* spectra

482 **figure 11:** examples of peaks extraction in *TNT* spectra acquired with *CdZnTe* detector (a) and *Ge*  
483 detector (b)

484 **figure 12:** two main peaks location of *Ge* spectra  $\theta=4.28^\circ$

485 **figure 13:** two main peaks location of *CdZnTe* spectra  $\theta=4.28^\circ$

486 **figure14:** visualization of a package containing an explosive substance

487 **figure 15:** visualization of the influence of attenuation on *Semtex* spectrum acquired with *Ge*  
488 detector when the transmitted signal is close to 65%

489 **figure 16:** influence of the attenuation on the substance identification with cross correlation  
490 coefficient

491 **figure 17:** detection algorithm principle

492 **figure 18:** illustration of the interface of the detection algorithm

493

494 **Table caption:**

495 **Table 1:** *substance identification using cross correlation coefficient*



496 **Table:**

497

<i>Material Tested</i>	<i>Germanium detector</i>	<i>CdZnTe detector</i>
<i>Aluminum</i>	Aluminum	Aluminum
<i>Coffee</i>	Coffee	Coffee <i>Cotton</i>
<i>Copper</i>	Copper	Copper
<i>Toothpaste</i>	Toothpaste	Toothpaste <i>PVC</i>
<i>Graphite</i>	Graphite	Graphite
<i>NaCl</i>	NaCl	NaCl
<i>Nickel</i>	Nickel	Nickel
<i>PVC</i>	PVC	PVC <i>Toothpaste</i>
<i>Sugar</i>	Sugar	Sugar
<i>Soap</i>	Soap	Soap
<i>Cotton</i>	Cotton	Cotton <i>Coffee</i>
<i>Semtex</i>	Semtex	Semtex
<i>Formex</i>	Formex	Formex <i>TNT</i>
<i>PLA NP</i>	PLA NP	PLA NP
<i>HCG</i>	HCG	HCG
<i>TNT</i>	TNT	TNT <i>Formex</i>

*Table 1: substance identification using cross correlation coefficient*

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