

Implementation of an imaging spectrometer for localization and identification of radioactive sources

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1	Implementation of an Imaging Spectrometer for Localization and
2	Identification of Radioactive Sources
3	
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7	•
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12	
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14	Telephone: 0033 1 69 08 35 75
15	A historia at
10 17	Adstract
17 18	Spatial localization of radioactive sources is currently a main issue interesting nuclear
19	industry as well as homeland security applications and can be achieved using gamma
20	cameras. For several years, CEA LIST has been designing a new system, called GAMPIX.
21	with improved sensitivity, portability and ease of use. The main remaining limitation of this
22	system is the lack of spectrometric information, preventing the identification of radioactive
23	materials. This article describes the development of an imaging spectrometer based on the
24	GAMPIX technology. Experimental tests have been carried out according to both
25	spectrometric methods enabled by the pixelated Timepix chip used in the GAMPIX gamma

camera. The first method is based on the size of the impacts produced by a gamma-ray energy deposition in the detection matrix. The second one uses the Time over Threshold (ToT) mode of the Timepix chip and deals with time spent by pulses generated by charge preamplifiers over a user-specified threshold. Both energy resolution and sensitivity studies demonstrated the superiority of the ToT approach which will consequently be further explored. Energy calibration, tests of different pixel sizes for the Timepix chip and use of the Medipix3 chip are future milestones to improve performances of the newly implemented imaging spectrometer.

33

34 Keywords

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36 Gamma imaging; GAMPIX; Timepix; imaging spectrometer; Time over Threshold

37 38

1. Introduction

39 Spatial localization of radioactive sources is currently a main issue interesting nuclear 40 industry (nuclear power plants security, decommissioning of nuclear facilities, radiation-41 protection) as well as homeland security applications (controls, post-accidental interventions) [1] [2]. Gamma imaging is a very interesting technique to achieve this spatial localization by
enabling superimposition of visible and gamma pictures using dedicated devices called
gamma cameras.

45 Spatial localization can be achieved using Compton scattering or coded masks. Compton approach includes two steps: the scattering of the incident photon and its full absorption. 46 47 From the path of each incident photons one can determine cones from which it could have 48 been emitted. The radioactive source is located at the intersection of all the rebuilt cones. Two 49 sensors are usually involved in these systems but gamma cameras based on a single sensor 50 also exist. We can give the example of the recently industrialized Polaris-H system [3] [4]. In 51 this new gamma camera, the depth of interaction required to determine the path is obtained 52 from the cathode-to-anode signal ratio (CAR) or from drift time information. Because photons 53 have to deposit energy in two successive detectors, the Compton approach is mainly dedicated 54 to photons above 200 keV [3]. In the rest of the article, we will focus on gamma cameras 55 using coded masks.

56 Current industrial gamma cameras based on coded masks can be considered as first generation 57 because they are based on scintillator detectors. Much progress was made since the design of 58 the first gamma camera by Hal Anger (Berkeley University, California) in the last's 50 for 59 medical applications [5]: digitalization of data processing [6], replacement of the pinholes 60 used for spatial localization (CARTOGAM, CEA LIST [7]) by multiple hole collimators 61 (Fixed Multiple hole Collimated Camera, University of Michigan [8]) and MURA coded 62 masks (RadCam, Radiation Monitoring Devices Inc. [9]), etc. In the 90's, continuous 63 scintillators moved into pixelated scintillators (CSPD-2, University of Michigan [10]; RMD-64 Pinhole, Radiation Monitoring Devices Inc. [11]). At the same time, semiconductor detectors were developed [6]. Such detectors intended to improve both spatial and energy resolution by 65 66 enabling direct conversion from gamma photons to electrical charge. First gamma cameras integrating semiconductor detectors present some limitations because of small detection 67 68 surfaces resulting in small fields of view ([12, 13]) and obligation of cooling the detector 69 when using materials such as germanium [12]. The progressive development of pixelated 70 CdTe or CdZnTe substrates hybridized to ASICs [14, 15, 16, 17] opened the way to a second 71 generation of gamma cameras operating at room temperature.

- 72 In this context, CEA LIST designed a second generation system, named GAMPIX [1, 18, 19].
- 73 GAMPIX's body integrates three main building blocks:
- 74 The detection system is a 1 mm thick CdTe substrate bump-bonded to a pixelated readout
- chip called Timepix [17] and developed by the CERN. In 1.4 cm², the Timepix chip integrates
- 76 256 pixels by 256 pixels, 55 μm side, with independent shaping and processing chains.
- 77 In front of the detection system, the coded mask in tungsten alloy is used as a multi pinhole
- collimator for spatial localization [20]. It is characterized by its number of holes (rank) and its
- 79 thickness.
- 80 Finally, the USB module enables plug-and-play connection of the gamma camera with the
- 81 acquisition laptop [21] and remote measurements.

- GAMPIX is currently under industrialization by AREVA CANBERRA (the industrial system 82 is named iPix, see Fig. 1). Compared to CARTOGAM, which is the current AREVA 83 84 CANBERRA industrial system, GAMPIX presents three main improvements:
- 85 The first one is the low-energy (below 100 keV) sensitivity with a gain of five decades in
- comparison with CARTOGAM. GAMPIX is able to detect in 1 s a ²⁴¹Am radioactive source 86
- generating a dose rate of $0.25 \,\mu$ Sv.h⁻¹ in the vicinity of the gamma camera. For this reason, 87
- GAMPIX is a performing tool for plutonium detection during decommissioning operations 88
- (²⁴¹Am being a feature of the presence of plutonium). GAMPIX efficiency decreases at high 89
- energy because of both the small detection volume (0.1982 cm³ of CdTe against 5 cm³ of 90 CsI(Tl) for CARTOGAM) and the non-perfect filtering achieved by the coded mask. For this
- 91 reason, 20 s are needed to detect a 137 Cs radioactive source with 2.5 μ Sv.h⁻¹ dose rate and 60 s 92
- 93 for a ⁶⁰Co source giving a dose rate of 3.8 µSv.h⁻¹. However, it is important to emphasize that,
- 94 by adapting the characteristics of the coded mask, GAMPIX is able to cover an energy range
- from ²⁴¹Am to ⁶⁰Co with better performances than CARTOGAM even at high energy (see [1] 95
- 96 for results obtained in Nuclear Power Plants).
- 97 The second point is the portability facilitated by the reduction of the weight. CARTOGAM,
- 98 which is the lightest system currently on the market, and GAMPIX respectively weight 15 kg
- 99 and 2 kg. The difference is mainly due to the shielding required by the scintillation detector of
- CARTOGAM. 100
- 101 Finally, the third point deals with the ease of use and deployment of GAMPIX in comparison
- 102 with CARTOGAM. GAMPIX uses for instance only one cable for camera management, data
- 103 transmission and power supply.
- 104 Besides, GAMPIX has a field of view of 50°. The angular resolution, which refers to the
- 105 minimal angle between two radioactive sources to be separated in the decoded image, reaches
- 106 down to 2° for a ²⁴¹Am radioactive source.
- 107 GAMPIX applications benefit from its characteristics. Thanks to its great portability, it can 108 easily be deployed in nuclear power plants in order to control, for instance, the correct 109 position of lead shielding dedicated to the radiation protection of operators. Regarding nuclear 110 facilities decommissioning, GAMPIX is able to provide an accurate localization of hot spots 111 (for instance, in pipes) for targeted decommissioning enabling both reduction of operations 112 duration and waste volume to be stored. The sensitivity of GAMPIX and its easy deployment 113 by non-expert end-users enable its use for fast control of luggage (airports) and containers 114 (ports) for homeland security applications. Finally, for post-accidental interventions, 115 GAMPIX can help the first responders to quickly identify dangerous areas in Fukushima type 116 environments. Experimental results illustrating these applications can be found in the
- 117 Reference [1].
- In its current version, the main limitation of the GAMPIX gamma camera is the lack of 118
- 119 spectrometric information, preventing the identification of radioactive material. Thus, dose
- 120 rate calculation needs an assumption on the nature of radionuclides and it is impossible to
- 121 identify different radionuclides simultaneously present in the environment. Considering this
- 122 limitation, it was decided to add new spectrometric capabilities to the GAMPIX gamma
- 123 camera to achieve an imaging spectrometer.

124 The Timepix chip offers two approaches for performing spectrometry measurements. The first one is based on the average size of the clusters which directly depends on the energy of the 125 incident gamma-ray. As an example, the average cluster size is contained for a given Timepix 126 energy threshold between 2.8 pixels for a ²⁴¹Am source and 7.0 pixels for a ⁶⁰Co source. The 127 incident average energy can thus be deduced from the average cluster size. The second 128 129 approach uses the Timepix Time over Threshold (ToT) mode [17, 22, 23]. By setting a 130 threshold on pulses obtained at the output of charge sensitive preamplifiers, ToT mode 131 measures the time spent by the pulses over the threshold, which is directly dependent on the 132 incident gamma-ray energy. Conversion between cluster sizes or ToT values and energy can 133 be achieved using reference radioactive sources or monoenergetic beams.

The purpose of this article is to demonstrate the ability of the GAMPIX system to provide spectrometric information. Qualitative and quantitative evaluation of its performances regarding this purpose will be presented. The first part of the document is dedicated to the preliminary setting of the Timepix chip and to the description of the required analysis tools. In the second part, methodology for implementing the imaging spectrometer and evaluation criteria of the final system are presented. Finally, the last part summarizes experimental results obtained in the frame of this study.

141

2. Settings of the Timepix chip and analysis tools

142 Fine tuning of the Timepix chip settings was crucial prior the implementation of the imaging 143 spectrometer. It aims at optimizing both energy resolution and gain. Settings and data 144 acquisition were performed using the Pixelman interface developed in the Czech Technical 145 University of Prague [24]. First, threshold equalization with "noise edge" method was carried 146 out to minimize dispersion around the average threshold value caused by gain differences 147 between pixels. Then, a parametric study on the thirteen chip parameters showed that the I_{krum} 148 DAC had the greatest influence on both energy gain and energy resolution [25]. The I_{krum} 149 current both controls falling times of pulses generated by charge preamplifiers and 150 compensates leakage currents (within the limit of $I_{krum}/2$). All parameters were finally set to 151 their default value, except Ikrum which was set to the DAC code value 2 corresponding to a 152 falling time in the order of 1 µs [26]. It is important to notice that the pile-up is limited with 153 such a value. The substrate bias voltage has to be high enough (in absolute value) to minimize 154 charge spreading and charge trapping which is a drawback of CdTe. In our case, bias voltage 155 was set to -110 V. Conversion between ToT values and energy can be done by mean of a calibration curve [27-28]. This curve is mostly linear, except at very low energy (just above 156 157 the threshold set on the pulses). Energy calibration also aims at optimizing energy resolution 158 by correcting the shift between peaks due to clusters of different sizes (Fig. 2). In this study, 159 we tested our imaging spectrometer without energy calibration but directly with ToT values. 160 It is important to emphasize on the fact that energy resolution improvement given by the 161 energy calibration step was not crucial for these measurements because gamma-ray spectra 162 coming from the different studied radionuclides have a typical signature (Table I).

Data processing was performed with dedicated MATLAB software developed by CEA LIST.
 This software implements processing functions dealing with both spectrometric approaches

165 tested in the imaging spectrometer. Concerning cluster size, the software identifies clusters as set of neighboring pixels. A maximal allowed cluster size can be set by the user to remove 166 167 cosmic rays, size of which commonly exceeds 20 pixels. It is important to set a low enough 168 acquisition time per frame to avoid pile-up which would lead to non-physical clusters 169 resulting from the sum of successive close events. Cluster size histograms giving the number 170 of occurrences depending on the cluster size are finally plotted. As far as ToT mode is 171 concerned, the software sums ToT values of all pixels forming a cluster. If energy calibration 172 has been achieved, energy conversion is done before summation. Spectra giving the number 173 of occurrences as a function of ToT values are finally plotted. The software also achieves 174 spatial reconstruction from the coded mask projection on the detection matrix. Spatial 175 reconstruction can be focused on a given cluster size windowing or ToT windowing specified 176 by the user. This functionality will be used for the implementation of the imaging 177 spectrometer as presented in section III.

178

3. Implementation of the imaging spectrometer and evaluation criteria

179 To demonstrate the feasibility of an imaging spectrometer based of the GAMPIX gamma camera, it was decided to develop a device achieving a selective spatial reconstruction 180 181 depending on the energy of incident photon (via cluster size and ToT values). This device was 182 tested with four radioactive sources covering the energy range of interest for the GAMPIX 183 system (see Table I). Performances of both spectrometric approaches in terms of 184 discrimination capability, and comparison with the GAMPIX gamma camera in its current 185 version in terms of sensitivity were assessed. The first part of this section is dedicated to the 186 methodology used for the implementation of the imaging spectrometer while the second part 187 justifies the choice of evaluation criteria.

188 **3.1 Implementation of the imaging spectrometer**

189 Implementation of the imaging spectrometer according to both cluster size and ToT values 190 approaches is based on windowing. Cluster size windowing requires a preliminary 191 measurement with each radionuclide taken alone. From the cluster size histograms, mean 192 cluster size, dispersion around the mean and overlapping between radionuclides are evaluated. 193 The first spatial reconstruction is performed on the single mean cluster size. Then, the 194 windowing is progressively broadened and the best configuration is determined by qualitatively evaluated spatial reconstructions. To appreciate differences between cluster size 195 histograms, Fig. 3 shows histograms of ²⁴¹Am and ⁶⁰Co radioactive sources and Table II gives 196 197 mean cluster size and percentage of clusters in different ranges for the four radionuclides 198 tested. Table III summarizes the cluster size windowing chosen for best discrimination. To 199 avoid overlapping between radionuclides, mean cluster size and most frequent cluster sizes 200 are not necessarily included in the windowing.

201 Concerning ToT windowing, a preliminary measurement with each radionuclide taken alone 202 is also required to identify in the spectra ToT values associated with typical features 203 (photoelectric peaks, Compton edge, etc.) Spectra obtained with each radionuclide are then 204 compared to determine if there is overlapping due to the energy resolution of the sensor. The 205 first windowing is centered on typical features and the best windowing is finally obtained by sequential approach. Typical features of the four radionuclides studies and position of the 206 207 windowing are shown in Fig. 4. All ToT spectra obtained are in good agreement with 208 previous literature results [27] and show the ability of the ToT mode to provide useful gamma-ray spectra, even at high energy (¹³⁷Cs and ⁶⁰Co). One can notice the large fraction of 209 210 events on the fluorescence and escape peaks. It is explained by the pixelation of the detector: 211 it is unusual that both incident photon and fluorescence photon deposit their energy in the 212 same 55-µm-side pixel. Table IV summarizes ToT windowing for all tested radionuclides. 213 Because of overlapping, windowing does not necessarily include typical features.

214 **3.2 Evaluation criteria**

The first evaluation criterion of imaging spectrometer performances is the discrimination ability, which is qualitatively evaluated from spatial reconstructions. If radionuclides with different gamma-ray emissions are simultaneously present in the field of view, the discrimination ability characterizes the ability of the system to reconstruct only radioactive sources included in a given energy range.

220 The second evaluation criterion is the sensitivity, which corresponds to the minimal duration 221 required for detecting a radionuclide inducing a given dose rate near the gamma camera. The 222 sensitivity corresponds to a picture free of artifacts, as shown in Fig. 5 (b). Three parameters 223 have a great impact on the sensitivity. The first one is the detector efficiency. It decreases 224 when the incident gamma-ray energy increases as shown in Fig. 6. For a 1-mm-thick CdTe 225 detector, efficiency drastically decreases from 100 keV. The second factor is the coded mask 226 and its characteristics. A tradeoff has to be found between thickness (sensitivity) and number 227 of holes (i.e. the rank, which defines the angular resolution). The last factor is the energy 228 windowing. Without windowing (standard working mode for the GAMPIX gamma camera), 229 all photons hitting the detector are taken into account. The narrower windowing is, the fewer 230 photons are considered, and the more sensitivity is decreased.

4. Performances of the imaging spectrometer

Performances according to both evaluation criteria presented in section III are successively
presented. All acquisitions were performed in "Time over Threshold" mode and in "frame"
type with 1 s acquisition per frame. They were repeated three times to control reproducibility.

235 **4.1 Discrimination ability**

To evaluate discrimination capability, radioactive sources were disposed two or three at a time in front of the gamma camera over a graduated table. Tests were carried out for distance between radioactive sources and GAMPIX gamma camera varying between 50 cm and 150 cm on the camera axis and between 0 and 50 cm on each side of the camera on the perpendicular axis. Acquisition time was set between 300 s and 2000 s depending on configurations tested. Table V shows pictures obtained after both cluster size windowing and ToT windowing for a 1500 s acquisition with ²⁴¹Am, ¹³³Ba and ¹³⁷Cs radioactive sources positioned in the configuration illustrated in Fig. 7. Both approaches are efficient for ²⁴¹Am and ¹³⁷Cs discrimination but cluster size windowing is unable to separate ¹³³Ba from ²⁴¹Am. ¹³⁷Cs also appears less punctual for cluster size configuration and there are more artifacts on ²⁴¹Am picture. All tested configurations proved the superiority of ToT approach on cluster size approach regarding this evaluation criterion.

Three factors explain this superiority. First, spectra of the four radionuclides tested are well differentiated contrary to cluster size plots: centroid of photoelectric peaks varies from about 500 to 4500 from ²⁴¹Am to ⁶⁰Co (Fig. 4), while mean cluster size only changes from about 3 to 7 (Table III). Secondly, the 11810 channels of the counting system in ToT mode are great enough to show these differences. Finally, ToT mode enables to carry out fine spectrometry measurements while cluster size mode only deals with mean energy values.

4.2 Sensitivity

256 During our experiments, sensitivity was determined for each source placed at 1 m from the

257 gamma camera in the camera axis, without windowing, with cluster size windowing and with

258 ToT windowing. Several configurations of the mask were tested. We were looking for the loss

- 259 of sensitivity induced by the spectro-imaging mode for both spectrometric approaches with 260 respect to the GAMPIX gamma camera in its current version.
- Tables VI to VIII summarize sensitivity for all radionuclides tested without windowing (GAMPIX gamma camera in its current version) and with cluster size and ToT windowing for several configurations of the coded mask. Percentages below the values indicate the loss of sensitivity due to both windowing techniques.
- Several conclusions can be drawn from these results. Concerning coded masks, the one of 265 rank 7 with a thickness of 4 mm produces best results for energies under 100 keV (²⁴¹Am, 266 267 ¹³³Ba), while mask of rank 7 with a thickness of 8 mm is more efficient for higher energies (¹³⁷Cs, ⁶⁰Co). Rank 13 offers better performances than rank 7 in terms of spatial resolution but 268 is less efficient in terms of sensitivity. Two millimeters appears to be a too-thin thickness for 269 270 each of the tested radionuclides, especially for high-energy gamma-ray emissions. In the case 271 of unknown searched radionuclides, coded mask of rank 7 with a thickness of 4 mm offers the 272 best tradeoff.
- 273 Concerning energy windowing, it causes a loss of sensitivity greater than 20% in most cases, 274 which is explained by the little fraction of events occurring in the sensor and finally selected for spatial reconstruction. For low energies, this sensitivity loss is not a real problem because 275 276 of very small acquisition times required by the GAMPIX gamma camera (from 1 s to 2 s for ²⁴¹Am with rank 7 and thickness of 8 mm for the coded mask). For higher energies, loss can 277 be limited by the choice of the most adapted mask. Best sensitivities are obtained for ToT 278 279 windowing in comparison with cluster size windowing. Degraded results with ⁶⁰Co are 280 explained by photoelectric peak spreading due to the high mean cluster size (7) and to the 281 dispersion around this value which causes shifts between photoelectric peaks.

282 **5.** Conclusion and outlook

283 The purpose of our study was to demonstrate the feasibility of an imaging spectrometer based 284 on the GAMPIX gamma camera and to evaluate its performances. Two methods were tested 285 to implement this imaging spectrometer: cluster size and ToT approaches. Tests on 286 discrimination ability and sensitivity both proved the feasibility of such a device and the 287 superiority of ToT approach. Loss of sensitivity with ToT approach is greater than 20%. If it 288 is not a problem for low energies, a relevant choice of the mask can mitigate this drawback 289 for energies higher than 100 keV. If the radionuclide is unknown, the coded mask of rank 7 290 with a thickness of 4 mm offers the best compromise.

291 Further developments of the imaging spectrometer will combine both cluster size and ToT 292 spectrometric approaches. Next planned step is the integration of the energy calibration in the 293 imaging spectrometer for the analysis of closer gamma-ray energies. Because of the high 294 mean cluster size, improvements are also expected for high energies (⁶⁰Co). Energy 295 calibration measurements should take place at the SOLEX facility which provides 296 monoenergetic beams from 0.5 keV to 28 keV [30]. As a first step, we plan the global calibration of the Timepix chip. Improvements of the energy resolution of a factor between 297 298 two and four are reported by [26] with a pixel by pixel calibration and this approach will be 299 considered as a second step. Test of a 1 mm thick, 110 µm pixel side Timepix chip is also 300 expected. It would enable to evaluate the energy resolution gain due to the limitation of 301 charge sharing between several pixels, which is one of the main explanations for energy 302 resolution degradation.

Finally, the replacement of the Timepix chip by a Medipix3 chip will be studied. The ToT mode is not implemented in the Medipix3 chip and spectra have to be obtained by counting the number of events for each threshold value [31]. The main improvement compared to previous Medipix chips concerns the hardware connection between several neighboring pixels, which should drastically improve the energy resolution of the system.

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379 Figures

380



- 381 Fig. 1: (a) GAMPIX gamma camera prototype developed by CEA LIST (b) front side and (c) back side of the
- 382 iPix industrial prototype developed by CANBERRA.

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384

385 Fig. 2: Spectra obtained with a 241 Am radioactive source depending on cluster size.

386



387 Fig. 3: Cluster size histograms for (a) 241 Am and (b) 60 Co







390



- 391 Fig. 5: Spatial reconstruction of a radioactive source of ¹³⁷Cs (coded mask of rank 7 with thickness of 4 mm) for
- acquisition time of (a) 10 s and (b) 400 s. The presence of artifacts can be observed on the left. Result obtained
- 393 for a 400 s acquisition time is considered as satisfying.









413	Table 4				
414		ToT windowing			
	R	adionuclide	Windowing (To	Γ values)	
		²⁴¹ Am	400-500)	
		¹³³ Ba	600-800)	
		¹³⁷ Cs	1000-200	00	
		⁶⁰ Co	5000-150	00	
415					
416			Table 5		
417	Mean cluster size and cluster windowing				
	Radioactive sources	All	241Am	133Ba	137Cs
	Cluster size window	ing	•	.	•
		- • •			

ToT windowing

418

419

Table 6

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420 Sensitivities without windowing and for cluster size and ToT windowing with rank 7, thickness of 4 mm coded 421 mask

	Without windowing	Cluster size windowing	ToT windowing
241 A m	1 s	1 s	1 s
AIII		0%	0%
133 D o	4 с	15 s	7 s
Da	4 8	+275%	+75%
137 C a	60 a	130 s	100 s
uncs.	60 S	+117%	+67%
60 C o	300 s	400 s	1500 s
C0		+33%	+400%

422 423

Table 7

424 Sensitivities without windowing and for cluster size and ToT windowing with rank 7, thickness of 8 mm coded 425 mask

mask			
	Without windowing	Cluster size windowing	ToT windowing
241 A m	1 s	2 s	2 s
AIII		+100%	+100%
133 D o	10 s	25 s	12 s
Da		+150%	+20%
137 C a	20 s	180 s	80 s
Cs		+800%	+300%
60 C o	60 s	125 s	650 s
		+108%	+983%

426

427

429

Table 8

430 Sensitivities without windowing and for cluster size and ToT windowing with rank 13, thickness of 2 mm coded 431 mask

	Without windowing	Cluster size windowing	ToT windowing
²⁴¹ A m	n 3 s	4 s	4 s
AIII		+33%	+33%
133 D o	14 c	100 s	17 s
Ба	14 8	+614%	+21%
1370-	200 -	> 600 s	> 600 s
.s,Cs	500 s	>+100%	>+100%
⁶⁰ Co	Not visible	Not visible	Not visible