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Classification-based material decomposition method for photon counting-based spectral radiography: application to plastic sorting

Ting Su^{a,1}, Valérie Kaftandjian^{a,*}, Philippe Duvauchelle^a, Yuemin Zhu^b

Abstract

Dual-energy X-ray radiography has been commonly used for materials separation. However, its performance is limited, especially for the separation of close materials or the identification of multiple materials. To cope with this problem, we propose to investigate the material decomposition ability of spectral radiography based on photon-counting detector. The latter having energy resolving capability can provide spectral information of several energy bins and thus enables selective imaging of multiple materials. In this framework, a classification-based patchwise regularized decomposition method was proposed to gain better differentiation between materials. It consists of performing several decompositions with reduced number of materials in the basis and classifying these decompositions using their cost function values. The results on simulations showed that, in the presence of Poisson noise, the method without classification can separate acrylonitrile-butadiene-styrene (ABS) from three kinds of flame retardants (FRs: brominated FR, chlorinated FR and phosphorus FR), but that the type of FR cannot be identified. With the classification technique, ABS and three kinds of FRs can be both separated and identified at the same time when the

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thickness was as large as 2 mm or 4 mm. The results on real data from physical photon-counting detector further confirm that the ABS, Br and Cl can be separated from each other.

Keywords: X-ray, radiography, photon-counting detector, plastic sorting

1. Introduction

X-ray radiography is an imaging technique that allows to view the internal structure of an object. Since the transmitted image is a 2-D projection of the 3-D object, useful information can be hidden because of physical overlapping. Dual-energy radiography using either different kVps or dual-layer detector is able to obtain two radiographs or images, one at higher energy and the other at lower energy. By making weighted subtraction of these two images [1, 2] or by performing a two-material decomposition [3], elimination of overlaying

materials and enhancement of the selective target material can be achieved.

- Dual-energy imaging has broad applications, such as rib suppression in chest radiography in medical domain [4, 5], threat detection in security inspections [6, 7, 8] and waste sorting in industrial applications [9, 10, 11]. However, its material discrimination ability is limited and materials with close atomic number (Z) can barely be separated.
- Thanks to the recent development of X-ray detector technology, a variety of pixelated photon-counting detectors (PCD) with imaging capability have been developped [12, 13, 14, 15]. By setting different thresholds (energy bins), each pixel of PCD is capable to either discriminate the transmitted photons corresponding to the selected energy bins, or firstly count the photons above the
- thresholds and then make subtraction to obtain the counts of each energy bin. With such spectral information, it is possible to set apart multiple components of the object with a single scanning and obtain improved performance in the aforementioned areas [16, 17, 18, 19, 20]. Most researches about the material decomposition ability of PCD concentrate on computed tomography (CT), called
- ²⁵ spectral CT. In the work of Schlomka *et al.* [21, 22], the measured attenua-

tion coefficients in spectral CT were decomposed into photoelectric absorption, Compton scattering and K-edge components, showing that dual contrasts can be well separated and quantified. Such K-edge technique is confined to the discrimination of high-Z materials with their K-edge inside the detection energy

- ³⁰ range. Other works [23, 24, 25, 26] considered the attenuation as a combination of several basis materials, whose distribution can therefore be quantified through decomposition process. Such approach is particularly suitable for the discrimination of lighter materials that have much lower K-edge energy beyond the detection energy range. In our previous work [27, 28], a patchwise regular-
- ized decomposition method based on basis materials combination was proposed for iron determination; the method allows iron to be quantitatively separated from calcium, potassium and water. However, potassium and calcium cannot be discriminated from each other since they have close Z numbers and hence close attenuation properties. The above material decomposition methods ini-
- tially proposed for CT can be applied to performing radiography, thus resulting in the so-called PCD-based spectral radiography named as spectral radiography in the following for simplicity. In the present work, we propose a new material decomposition method for spectral radiography and investigate its feasibility for plastic sorting.
- ⁴⁵ Waste electrical and electronic equipment (WEEE) has been increasing rapidly due to the development of electronic industry. In European Union, the amount of WEEE generated in 2005 is 9 million tons and this number is supposed to grow to 12 million by 2020 [29]. WEEE covers a wide variety of products such as lamps, hairdryers, computers, TV-sets, fridges and cell phones. This kind of
- ⁵⁰ waste contains various compositions among which polymers play an important part [30]. Acrylonitrile-butadiene-styrene (ABS) is a major component among all the polymers. Due to the presence of electronic power of EEE, the plastic materials should meet high fire safety standards. However, this is not possible to realize with pure polymers, therefore flame retardant (FR) are added to change
- ⁵⁵ flammability of plastics and increase the fire resistance [31]. Halogenated organic compounds (e.g., brominated aromatic compounds, chlorinated paraffins

and alicyclic compounds) and phosphorus compounds (e.g. organophosphates, halophosphates, phosphine oxides and red phosphorus) are two important FRs [32]. Recycling of plastics from WEEE is challenging because of the existence

- of FRs (especially the brominated and chlorinated FRs), which might result in serious environmental pollution. Therefore, sorting technique is important for subsequent treatment and recycling of WEEE to avoid major environmental and health problems. As mentioned before, some researches using dual-energy radiography for waste management have been reported. For example, Montag ner *et al.* [9] showed that heavy FR containing bromine (Br) is easier to be
- detected, but that chlorinated (Cl) FR can be sorted in the same family as Br, while the lighter one containing phosphor (P) remains undetectable when material thickness is less than 10 mm. To overcome these limitations, PCDbased spectral radiography appears an interesting solution due to its ability of decomposing multiple components of an object with a single scanning.

The purpose of the present work is to investigate the method of material decomposition for spectral radiography to discriminate plastic material from several FRs and further identify the type of FR. Based on our preliminary results reported in the ECNDT 2018 conference [33], the present paper aims to propose

⁷⁵ a new material decomposition method, which consists of performing several decompositions with reduced number of materials in the basis and classifying these decompositions using their cost function values. Both simulation and experimental results will be demonstrated.

This paper reuses a substantial part from one of the co-author's own thesis
⁸⁰ [28] with permission. More precisely, the reused contents appear mainly in sections I to IV and in Appendix.

2. Classification-based decomposition method

In our previous work [27], a patchwise regularized decomposition (PRD) method has been proposed, which divides the projection images into small patches and performs decomposition on each patch. The limitation of this method is to distinguish materials with close properties, and the performance of decomposition decreases when increasing the number of materials in the basis. To cope with the difficulty, the idea here is to perform several decompositions with reduced number of materials in the basis and classify these decompositions

⁹⁰ using their cost function values, thus leading to the so-called classification-based PRD (CPRD) method.



Figure 1: Flowchart of the decomposition method with classification for one patch. $fval_g$ represents the cost function value obtained by the gth decomposition with basis materials of *Group g*.

CPRD method also works in the way of patch-by-patch decomposition, but for several groups of materials. Figure 1 illustrates the procedure of this method for one patch. The principle is to firstly choose several groups of basis materials

- 95 (Group 1, Group 2, ..., Group g, ..., Group G), in which the same material can belong to several groups. Then, for each patch, we perform G times decomposition by carrying out PRD on each of basis material groups. For a Group g, the output is a set of decomposed density integrals at each pixel of the patch and a unique cost function value. After this step, we select the basis material group
- having the smallest cost function and we adopt the set of concentration values obtained from the decomposition based on this basis material group, while the

concentration of other materials inside this patch are set to 0. Finally, by repeating the above procedure for all the patches, we obtain the final image of each material.

105

More precisely, we consider the attenuation $\mu(\overrightarrow{x}, E)$ as a combination of M basis materials, where \overrightarrow{x} represents location and E represents energy. The basis materials can be chosen according to the prior knowledge of the scanned objects. We then have:

$$\mu(\overrightarrow{x}, E) = \sum_{\alpha=1}^{M} \rho_{\alpha}(\overrightarrow{x}) \mu_{m\alpha}(E), \qquad (1)$$

where $\rho_{\alpha}(\vec{x})$ denotes the density of material α at point \vec{x} and $\mu_{m\alpha}(E)$ the mass attenuation coefficient of material α at energy E. According to Beer-Lambert law, the expected number of photons λ_i , in energy bin $B_i(i = 1, 2...N, N)$ is the total number of energy bins) can be expressed as:

$$\lambda_{i} = \sum_{E=E_{s}(i)}^{E_{f}(i)} D(E)N_{0}(E)exp[-\int \mu(\overrightarrow{x}, E) ds]$$

$$= \sum_{E=E_{s}(i)}^{E_{f}(i)} D(E)N_{0}(E)exp[-\int \sum_{\alpha=1}^{M} \rho_{\alpha}(\overrightarrow{x})\mu_{m\alpha}(E) ds]$$

$$= \sum_{E=E_{s}(i)}^{E_{f}(i)} D(E)N_{0}(E)exp[-\sum_{\alpha=1}^{M} \mu_{m\alpha}(E)P_{\alpha}(sx, sy)], \qquad (2)$$

with

$$P_{\alpha}(sx, sy) = \int \rho_{\alpha}(\overrightarrow{x}) \, ds, \qquad (3)$$

where $E_s(i)$ and $E_f(i)$ denote respectively the start and final energies of bin B_i , D(E) is the detector absorption efficiency, $N_0(E)$ is the number of photons in the initial spectrum at energy E, and $P_{\alpha}(sx, sy)$ is the density integral that is line integral of object density $\rho_{\alpha}(\vec{x})$ along the measured projection path, with (sx, sy) representing the index of detector pixels.

To estimate $P_{\alpha}(sx, sy)$, we build an objective function that combines the log-least squares [34, 35], and a regularization term $R(P_{\alpha}^{C})$ for each small patch C on the attenuation image to reduce the effect of noise and enforce smoothness:

$$P_{\alpha}^{C}(sx, sy) = \arg\min_{P_{\alpha}^{C}(sx, sy)} \{ \sum_{(sx, sy) \in C} \sum_{i=1}^{N} [\ln(\lambda_{i}(P_{\alpha}^{C})) - \ln(m_{i}^{C})]^{2} + rR(P_{\alpha}^{C}) \},$$
(4)

where m_i^C is the measured number of photons in energy bin B_i and within patch C. r denotes the relaxation parameter. $R(P_{\alpha}^C)$ is the sum of the L2 regularizations of gradient images of P_{α}^C :

$$R(P_{\alpha}^{C}) = \sum_{\alpha=1}^{M} || \bigtriangledown P_{\alpha}^{C} ||_{2}^{2}$$

$$= \sum_{\alpha=1}^{M} \sum_{\substack{(sx,sy) \in C \\ (sx-1,sy) \in C \\ (sx,sy-1) \in C}} \{ [P_{\alpha}^{C}(sx,sy) - P_{\alpha}^{C}(sx-1,sy)]^{2} + [P_{\alpha}^{C}(sx,sy) - P_{\alpha}^{C}(sx,sy-1)]^{2} \}.$$
 (5)

3. Description of the spectral radiography simulation

125

We use the Virtual X-ray Imaging (VXI) software [36], which was developed in INSA Lyon, to simulate spectral radiography imaging. This software allows to define multiple parameters of the object to scan, including X-ray source, detector and system geometry.

Table 1: Components of three ABS-FR materials used for the phantom.						
Material(ABS-FR)	ABS-	ABS-	ABS-			
	TBBPA	DDC-CO	RDP			
Density of material $\rho(mixture)$	$1060~{\rm mg/cm^3}$	$1060~{\rm mg/cm^3}$	$1060~{\rm mg/cm^3}$			
Chemical formula of FR	$\mathrm{C_{15}H_{12}Br_4O_2}$	$\mathrm{C}_{18}\mathrm{H}_{12}\mathrm{Cl}_{12}$	$\mathrm{C}_{30}\mathrm{H}_{24}\mathrm{O}_8\mathrm{P}_2$			
Mass % (ω) of FR	15%	15%	15%			
Mass % (ω) of Br, Cl and P, respectively	8.82%	9.76%	1.62%			
$\rho_{\rm eff}$ *of Br, Cl and P, respectively	$93.5 \ \mathrm{mg/cm^3}$	$103.5~{\rm mg/cm^3}$	17.2 mg/cm^3			

*: ρ_{eff} is the effective density calculated by $\rho_{\text{eff}}(\alpha) = \rho(\text{mixture}) \times \omega(\alpha)$.

For example, $\rho_{\text{eff}}(\text{Br}) = \rho(\text{mixture}) \times \omega(\text{Br}) = 1060 \times 8.82\% = 93.5 \text{ mg/cm}^3;$

 $\rho_{\rm eff}({\rm ABS}) = \rho({\rm mixture}) \times \omega({\rm ABS}) = 1060 \times (1 - 15\%) = 901 \ {\rm mg/cm^3}.$

3.1. ABS-FRs phantom



Figure 2: Illustration of the ABS-FR phantom used for spectral radiography imaging. Materials in the cubes are given in Table 1.

We simulate a phantom containing multiple polymer+FR mixtures. As men-130 tioned above, ABS is a major component among the polymers used in electrical and electronic equipments. Therefore, we selected it as the polymer material for investigation. We also chose three kinds of commonly used flame retardants, including the brominated, chlorinated and phosphorus FRs. They are respec-

135

tively tetrabromobisphenol A (TBBPA), dechlorane plus (DDC-CO) and resorcinol bis(diphenyl phosphate) (RDP). Three ABS-FR materials were obtained by mixing each FR with ABS at mass percentage of 15%. Detailed information of these materials is summarized in Table 1.

The simulated phantom is composed of multiple cubes with height of 10 mm, width of 10 mm and different thicknesses. As shown in Figure 2, each column of cubes are of the same material (denoted on the top) and each row of cubes are of the same thickness (denoted on the left). Figure 3 plots the mass attenuation coefficients of the components contained in the phantom: ABS, Br, Cl and P, which will be used to distinguish different ABS-FR materials.



Figure 3: Mass attenuation coefficients ($\mu_{\rm m}$) of components (ABS, Br, Cl and P) within the detection energy range from 30 keV to 90 keV. $\mu_{\rm m}$ of ABS is calculated according to formula $\mu_{\rm m} = \sum_{\alpha} \omega_{\alpha} \mu_{\rm m\alpha}$, with ω_{α} and $\mu_{\rm m\alpha}$ representing the weight fraction and mass attenuation coefficient of each element α of the compound. Data taken from [37].

145

From the decomposition method described in the above, the finally obtained P_{α} for material α is the line integral of material density ρ_{α} (Equation 3). In this case, with known thickness d of each cube, theoretical values for P_{α} at position

Table 2: Theoretical values of P_{α} for each material. Material ABS has two columns: the first is *ABS pure*, which corresponds to the material of the 4th column of cubes in the phantom shown in Figure 2, and *ABS in mixture* corresponds to the ABS compound in the other cubes of Figure 2.

Thickness ABS pure (mm) (mg/cm ²)	ABS in	Br in	Cl in	P in	
	ADS pure $(, 2)$	mixtures	ABS-TBBPA	ABS-DDC-CO	ABS-RDP
	$(\mathrm{mg/cm^2})$	$(\mathrm{mg/cm^2})$	$({\rm mg/cm^2})$	$(\mathrm{mg/cm^2})$	
0.3	31.80	27.03	2.81	3.11	0.52
0.5	53.00	45.05	4.68	5.18	0.86
1	106.00	90.10	9.35	10.35	1.72
2	212.00	180.20	18.70	20.70	3.44
4	424.00	360.40	37.40	41.40	6.88

(sx, sy) of the transmitted image is given by

$$P_{\alpha}(sx, sy) = d(sx, sy) \times \rho_{\alpha}(sx, sy), \tag{6}$$

where ρ_{α} equals the effective density calculated in Table 1. Table 2 lists the theoretical values of P_{α} for materials ABS, Br, Cl and P at various thicknesses.

3.2. Acquisition parameters and system geometry



Figure 4: Scheme of spectral radiography system geometry. This figure is only a simplified schematic view of the simulation system where the dimensional information $(D_{SC}, D_{DC}, \text{size})$ of phantom and detector) is not in real scale.

The simulated system used a point X-ray source of tungsten target material and the target angle was 17°. According to reference [38], tube voltage was set to be 100 kVp, tube current 15 mA and exposure time 1 s. The X-ray spectrum

- was computed based on Birch & Marshall model [39] without filtration. A 90×112 CdTe detector array with pixel size of 0.5 mm \times 0.5 mm and thickness of 3 mm was simulated. Six energy bins were set to be evenly distributed from 30 keV to 90 keV. We assumed that the detector has ideal energy resolution, detector absorption efficiency was simulated as the fraction of photons absorbed
- ¹⁶⁰ by the detector material. The distance from X-ray source to phantom center was 2000 mm and the distance of detector to phantom center was 3 mm. Figure 4 is a schematic view of the simulated spectral radiography system. Photonic noises were simulated by simple Poisson distribution with means equaling the expected numbers of photons acquired for each energy bin [40, 41]. This process was implemented in Matlab using command *poissrnd*.

4. Simulation Results

We have simulated two sets of spectral radiography acquisition data: with and without photon noise. To better evaluate the performance of the proposed CPRD method, it was also compared with PRD method without classification.

170 4.1. Decomposition results of PRD method

Regarding the components of the phantom, it is normal to consider a four basis material decomposition: ABS, Br, Cl and P. However, our experiment results show that P and Cl can barely be separated from each other since they have too close atomic number and hence close attenuation properties. Therefore,

we decompose the radiographic images into three basis images corresponding to ABS, Br and Cl and expect that P will also be present in the Cl basis image. The patch size is set to be 2×2 pixels.

For simulated acquisition without noise, the relaxation parameter r is set to 0 since it is not necessary to enforce smoothness within patches without noise. The decomposition results are shown in Figure 5(a). It is observed that:



Figure 5: Decomposition results (P_{α}) under different conditions using PRD method. (a): simulation without noise, r = 0; (b): simulation with Poisson noise, r = 0; (c): simulation with Poisson noise, $r = 2.51 \cdot 10^4$. The first column represents ABS basis, second column Br basis, and third column Cl basis image. The colorbars indicate decomposed P_{α} values in unit of mg/cm², whose true values are given in Table 2. The dash lines on images of (a) mark the position where 1-D profiles are plotted in later analysis.

- In the ABS basis image, all cubes are visible as expected, because ABS is present in both mixtures and pure ABS.
- In the Br basis image, the ABS-TBBPA cubes are well separated and highlighted with the P_{α} close to the true values, and no other FRs are present.
- In the Cl basis image, all the ABS-DDC-CO cubes are well highlighted as expected. At the same time, ABS-RDP is identified as Cl as expected, with P_α close to true values. However, for small thicknesses (d ≤ 2 mm), a slight amount of ABS-TBBPA is misidentified as Cl.
- Thus in the noise-free situation, ABS and Br can be well separated and identified. The determination performance of other FRs improves with the increase of object thickness: when d = 4 mm, the FRs containing Cl and P are present in the Cl basis image without cross-talk with the ABS-TBBPA material, one can further distinguish the two FRs according to their large density difference. In other words, if the thickness is known, the density can be used to identify the right material.

When Poisson noise is added, we need to reconsider the relaxation parameter r. r is determined as $2.51 \cdot 10^4$ in this situation according to the L-curve criterion. Some details of r selection using the L-curve method are introduced in the Appendix The decomposition results when r = 0 and $r = 2.51 \cdot 10^4$ are shown in Figure 5(b) and (c). It can be seen that when r = 0 (without regularization), the ABS-TBBPA cubes and ABS-DDC-CO cubes appear in both Br basis image and Cl basis image, making them impossible to be distinguished. Moreover, the decomposed images suffer from heavy noise. When the relaxation parameter was

set to $r = 2.51 \cdot 10^4$, the obtained images are less noisy than those in the former case, however, the separation of different materials is not enhanced. Another method is thus necessary.

4.2. Decomposition results with the new CPRD method

Instead of the 3-material (ABS, Br and Cl) decomposition using PRD method, we perform two independent decompositions with two groups of basis materials, group 1 is ABS + Br, and group 2 ABS + Cl. We give up a third group of ABS + P because it generates comparable cost function values with group 2, resulting in severe cross-talk between Cl and P. With the selected two groups of decomposition, by comparing the cost function values (fval1 and fval2) at

- the end of each decomposition, one of the two decompositions is chosen for a given patch. For example, if fval1<fval2, we choose the results of the decomposition that decomposes the data into ABS and Br, consequently, the values of Cl in this patch will be set to 0. After all patches being considered, full images of ABS, Br and Cl are obtained. Figure 6 shows the decomposition results of</p>
- ²²⁰ CPRD method and a reference method in the same condition as that in the above method without classification. The reference method employs a widely used least squares error objective function to solve the decomposition problem pixel by pixel [42].



Figure 6: Decomposition results under different conditions using CPRD method and reference method: (a) CPRD method, simulation without noise, r = 0; (b) CPRD method, simulation with Poisson noise, r = 0; (c) CPRD method, simulation with Poisson noise, $r = 2.51 \cdot 10^4$; (d) reference method, simulation with Poisson noise. The first column represents ABS basis, second column Br basis, and third column Cl basis image. Each column shares the same colorbar which indicates decomposed P_{α} values in unit of mg/cm², whose values are given in Table 2. The dash lines on images of (a) mark the position where 1-D profiles are plotted in later analysis.

In the noise-free condition (Figure 6(a)), FRs containing ABS and Br are identified respectively by CPRD method in the ABS and Br basis images as expected. Meanwhile, only the cubes of FR materials containing Cl and P appear in the Cl basis image, but they have significant density difference for cubes with the same thickness, therefore, they can be easily distinguished by the observer even if the concentration of FRs or thickness of cubes changes more or less.

In the presence of Poisson noise, Figure 6(b) shows the decomposition results of CPRD method when r = 0 (without regularization). Noise can be observed in decomposition images, however, material separation is better than with PRD method (compared with Figure 5): only few pixels containing Cl are misidentified in the Pr basis image and vice verse. The mass talk situation

- are misidentified in the Br basis image and vice versa. The cross-talk situation improves with the increase of thickness. In Figure 6(c), when $r = 2.51 \cdot 10^4$ (with regularization), the material separation is further enhance to some extent compared with Figure 6(b): less pixels of ABS-DDC-CO cubes appear in the Br basis image, and only the ABS-DDC-CO and ABS-RDP cubes are highlighted
- in the Cl image when d = 2 mm or 4 mm, like the results in noise-free case. In contrast, the reference method fails to separate Br and Cl. As can be seen in 6(d), the cubes containing Br also appear in the Cl basis image, no matter what thickness the sample has. Therefore, the CPRD method outperforms the reference method for material decomposition.

A more quantitative comparison is done in Figure 7, where the profiles of computed P_{α} with PRD and CPRD method are plotted together with the theoretical values (along dash lines in both Figure 5 and 6). It is observed that the CPRD method yields closer results to true values with respect to the PRD method especially for the Cl basis curve, where ABS-TBBPA cubes are mistaken

as Cl containing FR by PRD method. It is noteworthy that the theoretical values of P_{α} for the ABS-RDP cube in the Cl basis image are set to 1.72mg/cm^2 (corresponding to Table 2, the P_{α} of P in ABS-RDP with thickness of 1 mm) instead of 0, because we expect that P will appear in the Cl basis image.

So, the CPRD method has better decomposition ability than PRD for the

sorting application, no matter when Poisson noise is or is not present. ABS and Br containing FR can be identified in their corresponding basis images. Furthermore, when the cube thickness is increased to 2 mm or 4 mm, the decomposition results of Poisson noise condition become much closer to those of noise-free condition, where ABS and the FRs containing Br, Cl and P can be the use the use the second second





Figure 7: Performance comparison of PRD and CPRD methods in noise-free condition: 1-D profiles along the dash lines in Figures 5 and 6. ABS basis (top), Br basis (middle) and Cl basis (bottom). Black curves with point marker represent the theoretical density integrals of basis materials (P_{α} for thickness of 1 mm in Table 2)for perfect decomposition. Blue curves with triangle marker represent the calculated values using PRD method and the magenta curves with cross marker using CPRD method.

5. Preliminary experimental results

In order to evaluate the performance of our proposed method on physical data, we carried out radiography measurements using a CdTe photon-counting detector from Detection Technology company. The detector has linear array

²⁶⁵ structure with 256 detector pixels, each pixel is of 0.8 mm pitch size. The detector provides the possibility of setting 64 uniformly distributed energy bins between 20 keV and 160 keV. Considering our tube voltage of 110kVp for this experiment, we used data of the first 36 bins (20 keV to 110 keV) to perform material decomposition. As shown in Figure 8, the object being scanned is ²⁷⁰ composed of 5 materials: polyvinyl chloride (PVC), ABS with 20% Br, ABS with 5% Cl, polypropylene (PP) with 20% CaCO₃, and ABS pure. Acquisition parameters were set as: tube current 200 μ A, exposure time 100 ms, source to object distance 46 cm and object to detector distance 13 cm.



Figure 8: Illustration of experimental specimen (left) and system geometry (right).

Because of the linear structure of the detector, we have acquired single line data of 256 pixels. Therefore, the patchwise regularization, which requires 2 dimensional gradient, can not be directly applied. We hence adjusted our CPRD method by removing the regularization term and using patch size of 1x1 for experimental validation. 3 groups of basis materials were selected: group 1 "ABS + Br", group 2 "ABS + Cl", group 3 "ABS + Ca".



Figure 9: Decomposition results of ABS (a), Br (b), Cl (c) and Ca (d). The green check mark indicates a good decomposition while the red cross mark indicates a false decomposition.

The decomposition results are shown in Figure 9. It can be observed that all 5 materials are visible in Figure 9(a), since they either really contain ABS or contain components like PVC or PP that have similar attenuation properties to ABS. The pure ABS curve appears only in Figure 9(a), which indicates a good separation from other basis materials. In Figure 9(b), component Br is well discriminated. In Figure 9(c) the Cl component in PVC is well identified. However, a mistaken peak of PP with 20% CaCO₃ also appears, indicating that Ca probably cannot be separated from Cl. Also, the sample containing 5% Cl remains undetected.

In short, the proposed method achieves to identify ABS, Br as well as Cl with relatively higher concentration, but materials with close atom numbers, such as Ca and Cl remain unseparated.

6. Discussions

We have proposed a classification-based material decomposition method, which achieves to identify several different plastic materials in both simulation ²⁹⁵ and experimental validations. There are several parameters and factors that may influence the decomposition performance as discussed below.

Choice of basis materials. During simulation, we have found that using basis materials that have close chemical formulas to those actually present in the object can improve the decomposition performance. For example, using the same simulation data with Poisson noise, besides the above presented results, we have also performed a 3-group decomposition with basis materials like, *Group 1* (ABS + TBBPA), *Group 2* (ABS + DDC-CO) and *Group 3* (ABS + RDP). The results showed that with cube thickness of 2 mm and 4 mm, ABS and three FRs can be identified according to their corresponding decomposition image. That

is to say, DDC-CO and RDP can be distinguished from each other directly by decomposing them into their corresponding basis, instead of by their density difference in the same basis (Cl) image, as described in the above results of the CPRD method. We have not chosen these groups of basis materials with better performance because they are too specific. However, for the application where
the components of object are well known, appropriate choice of basis materials can yield better decomposition performance.

Patch size. In the present study, we have used a patch size of 2 × 2 pixels. To investigate the influence of patch size on decomposition performance, we plot in Figure 10 the curve of quantification error versus patch size, where quantification error represents root-mean-square-error (RMSE) of decomposed Br basis image. As observed, 2 × 2 patch size gives the smallest RMSE. Quantification error increases when patch size is larger than 2. This could be caused by the fact that large patch size results in over-smoothed image [43]. As a result, since there exist sharp grid-like edges throughout our entire image, over-smoothing
of these edges leads to larger quantification errors.



Figure 10: RMSE of quantified Br densities versus patch size.

Relaxation parameter r. Although the separation between materials is not enhanced (for PRD method) or slightly enhanced (for CPRD method) by choosing r according to L-curve method, it is still meaningful that noise (indicated by the variation of pixel values) of decomposition image can be decreased, which is obvious in Figure 5. We have applied L-curve method to certain region of the radiographic images, and used the same r for all patches. However, r should perhaps be changed for different patches if the signal-to-noise ratio varies. Therefore, further improvement could be achieved by tuning the relaxation parameter in each patch.

- Placement of energy bins. In the present work, we have used 6 uniformly distributed bins within detecting energy range to evaluate the performance of our proposed method in a general way. However, according to some researches [44, 45], appropriate selection of energy bins may have large influence on decomposition performance, especially when the total number of bins are small. In
- Figure 11(b), we demonstrate the decomposition results using a different strategy to place 6 energy bins. More bins were placed in lower energy on purpose since lower energy carries more material-differentiation information. We can observe performance improvement compared to Figure 11(a). From the ROI marked by the red circles, Br was better recognized with less cross-talk in Cl
- basis. Moreover, the calculated RMSE of Br basis image decreased from 1.23 to 0.88, indicating less quantification errors.



Figure 11: Comparison of decomposition results using different strategies of selecting energy bins. (a) 6 energy bins are uniformly distributed between 30keV and 90 keV, which is the same as Figure 6 (c). (b) More bins were placed in lower energy range as follows: 30 - 34, 35 - 41, 42 - 50, 51 - 61, 62 - 74 and 75 - 90 keV.

Detector response. In the present work, simulation validation only considers the absorption efficiency of the detector. However, in reality, photon counting detector's response function can be modeled by mixture of Gaussian functions allowing taking into account several artifacts such as electronic noise, 345 charge sharing, pulse pile-up, K-escape and Compton scattering [46]. Electronic noise is an important factor that limits the energy resolving capability of detector, it results in spectral resolution degradation and potential false counts especially at low energy. [47]. Charge sharing effect occurs when the charge cloud generated from X-ray interaction is shared by neighboring pixels and re-350 sults in two or more counts in several pixels and with lower energies. This effect is especially serious when pixel size is less than 0.5 mm [48]. Pulse pileup takes place at high count rate; two nearly simultaneously incident photons are counted as one, leading to loss of count and wrong recorded energy. This effect can be reduced by controlling incident count rate [47]. K-escape effect 355

is due to the characteristic X-rays induced by photoelectric effect. It can lead to different consequences: the recorded energy is lowered by K-shell energy, or two counts are generated, or still pile-up effect is induced [49].Compton scattering refers to Compton effect-induced scattered photons that are detected by

- an adjacent pixel or leave the PCD completely, resulting in long tail in lower energy. All these effects lead to distorted detector response and reduction of energy resolution. The degraded performance of material decomposition caused by discrepancy between theoretical and measured data can be addressed by two approaches. The first one is to characterize detector response by synchrotron
- radiation, radioactive isotopes or x-ray fluorescence [50], and then incorporate this information into material decomposition [51]. The second one is to calibrate material's linear attenuation coefficient by scanning samples with known components and thickness, so as to take into consideration the influence of detector response. The second approach is more flexible, easy to accomplish and efficient according to our preliminary test on experimental data. Though this strategy was not used for the demonstrated results here, we can still observe pretty good decomposition performance, indicating that our proposed method is able to process real experimental data.
- There are certain limitations of the proposed method. Firstly, the CPRD ³⁷⁵ method is not enough robust to noise when the object thickness is small. Secondly, the decomposition of the two FRs with similar properties (DDC-CO and RDP), even in the noise-free case, depends on their density difference in the Cl image. Therefore, in case where the thickness information of objects is unknown, or the two materials overlap in the X-ray direction, the decomposed density will be largely influenced, yielding the materials indistinguishable. Although an optional solution has been mentioned in the above, which consists in using directly DDC-CO and RDP as basis materials, but in a more generalized situation, close materials are still hard to be distinguished. Thirdly, the proposed CPRD method performs several decompositions into different groups of
- basis materials and then uses a classification technique to decide the right type of material. Although a decreased number of basis materials in each group re-

duces the running time of each decomposition, compared with a normal one with multiple basis materials, but on the other hand, the computation time can be increased because of extra decomposition procedures. For instance, in the present study, the 2-group (two basis materials in each group) CPRD decomposition

has about 36% longer running time than the 3-material PRD decomposition.

390

7. Conclusions

To investigate the ability of PCD-based spectral radiography for plastic sorting, we have proposed a classification-based decomposition method and evalu-³⁹⁵ ated its performance through both simulation and experimental validations. The simulation results showed that the proposed CPRD method is able to distinguish ABS and the FRs containing Br, Cl and P at the same time if the cube thickness is as large as 2 mm or 4 mm. Moreover, the quantified material densities agree well with theoretical values. Experimental validations further ⁴⁰⁰ confirm that ABS, Br and Cl can be separated from each other, but that materials with close atom numbers, i.e. Ca and Cl remain unseparated. Despite some limitations, our results suggest the possibility that PCD-based spectral radiography can be used to sort different types of plastic materials.

In addition to plastic sorting presented in the present work, the proposed ⁴⁰⁵ method can be readily applied to other applications such as industrial and medical ones. For example, in explosive detection, the explosive substances have close X-ray attenuation properties with some common materials such as sugar and polyethylene, a spectral radiography scan followed by the CPRD decomposition may improve their separation. Furthermore, the proposed method can

⁴¹⁰ be adapted to spectral photon-counting CT by adding a reconstruction step to transform the projection density integral information into cross-sectional density distributions. Thus spectral CT inspections could benefit from the proposed method too, such as abdominal imaging and atherosclerosis imaging, where material-specific imaging is of great importance for characterizing lesions or ⁴¹⁵ high risk plaques.

Appendix A. Determination of r with L-curve method

The L-curve is a log-log plot of the norm of a regularized solution versus the norm of the corresponding residual, where the "corner" of the curve corresponds to the selected relaxation parameter [52]. The solution and residual norms correspond to the objective function value and $R(P_{\alpha}^{C})$ value in Equation 4. We choose a region of interest on the radiographic image for the determination of r, which is shown in Figure A1.



Figure A1: Illustration of ROI for the determination of r.

The ROI contains 20×20 pixels. Given that the patch size is 2×2 , there are $10 \times 10=100$ patches within the ROI. For a given r, we define the solution norm and residual norm as the average values calculated over the 100 patches. We plot the L-curve with r varying from 10^{-10} to 10^{10} , as shown in Figure A2 (a). It is observed that the "corner" exists between $r = 10^4$ and $r = 10^5$. Furthermore, by plotting the same curve for a set of r between 10^4 and 10^5 , we have the curves shown in Figure A2(b). From this figure, we select $r = 2.51 \cdot 10^4$ as the relaxation parameter for all patches.



Figure A2: The L-curve as a function of residual norm for different values of the relaxation parameter r (a), and a magnified view of the blue circle region, where r varies from 10^4 to 10^5 (b).

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