


A nanomaterial release model for waste shredding using a Bayesian belief network

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Abstract The shredding of waste of electrical and electronic equipment (WEEE) and other products, incorporated with nanomaterials, can lead to a substantial release of nanomaterials. Considering the uncertainty, complexity, and scarcity of experimental data on release, we present the development of a

Bayesian belief network (BBN) model. This baseline model aims to give a first prediction of the release of nanomaterials (excluding nanofibers) during their mechanical shredding. With a focus on the description of the model development methodology, we characterize nanomaterial release in terms of number, size, mass, and composition of released particles. Through a sensitivity analysis of the model, we find the material-specific parameters like affinity of nanomaterials to the matrix of the composite and their state of dispersion inside the matrix to reduce the nanomaterial release up to 50%. The shredder-specific parameters like number of shafts in a shredder and input and output size of the material for shredding could minimize it up to 98%. The comparison with two experimental test cases shows promising outcome on the prediction capacity of the model. As additional experimental data on nanomaterial release becomes available, the model is able to further adapt and update risk forecasts. When adapting the model with additional expert beliefs, experts should be selected using criteria, e.g., substantial contribution to nanomaterial and/or particulate matter release-related scientific literature, the capacity and willingness to contribute to further development of the BBN model, and openness to accepting deviating opinions.

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Introduction

Rapidly developing markets such as green construction, energy harvesting and storage, advanced materials for aerospace, electronics, medical implants, and environmental remediation are potential key application targets for nanomaterials. There, nanotechnology has the potential to make qualitative improvements or indeed even to enable the technology (Kearney 2017). Impacts range from increased efficiency of energy harvesting or storage batteries to radical improvements in mechanical properties for construction materials. In addition, concerns of these markets such as scarcity of materials, cost, security of supply, and negative environmental impact of older products could also be addressed by new nano-enabled materials. For this, the development of a novel framework to enable naming, classification, hazard, and environmental impact assessment of present and future generation nanomaterials (Renn and Roco 2006) is a prerequisite for their safe, sustainable, and responsible industrial development (FutureNanoNeeds 2017; Lynch 2014).

FutureNanoNeeds, a EU seventh framework project, aims to do so by primarily responding to regulatory needs of future nanomaterials and markets. This project integrates concepts and approaches from several well-established contiguous domains to develop a robust, versatile, and adaptable naming approach, coupled with a full assessment of all known biological protective responses as the basis for a decision tree for screening exposure and hazard of nanomaterials at all stages of their life cycle. Together, these tools form the basis of a value chain (VC) regulatory process which allows each nanomaterial to be assessed for different applications on the basis of available data and the specific exposure and life cycle concerns for that application.

A material or substance flow analysis is a useful technique to quantify potential emissions of various substances, including nanomaterials, into different environmental compartments like air, surface or ground water, sediments, etc. (Kaegi et al. 2008; Arvidsson et al. 2011; Gomez-Rivera et al. 2012; Gottschalk et al. 2015). The potential sources of emission in product's different life cycle stages are first estimated in this technique. A mass balance is then done between the amount of nanomaterial present in the product and its emitted percentage for each estimated life cycle stage. Hauck et al. (2017) used this technique to quantify potential lead emissions from production, use, and end-of-life of perovskite in tandem solar cells. They

found end-of-life stage to be the main contributor to the emission of lead into the environment. Within the framework of FutureNanoNeeds, a prior material flow analysis was also carried out, as mentioned by Hauck et al. (2017), for several VCs involving perovskite-based photovoltaic (PV) panels, lithium ion batteries, and quantum dot-enabled electronic displays. The shredding of the waste generated from these VCs, i.e., waste of electrical and electronic equipment (WEEE) (Mitrano et al. 2015), was identified as one of the hotspots of nanomaterial emissions among other life cycle stages—an observation similar to the findings in the pertinent literature (Bystrzejewska-Piotrowska et al. 2009; Ling et al. 2012; Marcoux et al. 2013). Other findings in the literature include Caballero-Guzman et al. (2015) who found the recycling stage to be a “hotspot” of nanomaterial exposure during the assessment of flows of nanoparticles of TiO₂, ZnO, Ag, and CNT in the recycling system in Switzerland. Deng et al. (2014) found a high concentration of heavy metals (Cu, Pb, Cd, Cr, and Ni) in the sampled dust during the exposure assessment of shredding at a WEEE recycling site. The findings of Kohler et al. (2008) suggested the likeliness of the release of CNTs during disposal phases of CNT-treated products like Li-ion batteries and textiles.

In spite of the concern with nanomaterial release/exposure during shredding of nano-enabled products, the experimental data is uncertain, complex, and sparse (Chien et al. 2003; Oguchi et al. 2012; Part et al. 2015). In this context, the use of a Bayesian belief network (BBN) to forecast the nanomaterial release can be useful. BBN represents a branch of Bayesian modeling where the probability distributions are generally expressed in discrete form and solved analytically (Uusitalo 2007). A BBN is a directed acyclic graph that provides a coherent structure to make a priori assumptions about unknown variables, which can be used to generate forecasts and associated levels of uncertainty. It is expected to be advantageous in the forecasting of nanomaterial release because it can handle missing data, facilitate the learning of causal relationships between variables, and show good prediction accuracy also with smaller sample sizes; they also consist of formal rules that can be updated when new information becomes available (Wiesner and Bottero 2011; Uusitalo 2007). In terms of complexity, BBN has been shown to be pragmatic and scientifically credible to model complex and uncertain systems (Marcot 2012; Money et al. 2012;

Beaudrie and Kandlikar 2011). More interestingly, it has been explicitly used in the past to assess the risks associated with nanomaterials to both humans and environment (Money et al. 2012; Marvin et al. 2017; Bilal et al. 2017; Murphy et al. 2016). For example, Money et al. (2012) developed a baseline BBN model that integrates Ag nanoparticle-specific characteristics and aquatic environmental parameters for forecasting their fate and risks. Marvin et al. (2017) predicted biological effects and hazard potential of TiO₂, SiO₂, Ag, CeO₂, and ZnO nanoparticles in humans. Bilal et al. (2017) assessed multimedia distributions and concentrations of Al₂O₃, CeO₂, Cu, SiO₂, TiO₂, and ZnO nanoparticles in the environment by using a BBN model. Murphy et al. (2016) mapped the secondary data on the occupational exposure, obtained from US NIOSH exposure recommendation reports and several EU-funded research projects based on the risk estimation of CNTs, Ag, and TiO₂ nanoparticles to a control banding using BBN. One common limitation of these studies is that they consider nanomaterials in their individual or agglomerated form upon release. However, in reality, the released nanomaterials are normally found attached to or embedded in the matrix of the product from which they were released (Lowry et al. 2010; Shandilya et al. 2014a).

This article presents a baseline model which aims to give a first forecast of the potential airborne releases of nanomaterials during shredding for three VCs involving perovskites, quantum dots, and carbon-based nanomaterials. Instead of developing a model which is based on nanomaterial specific properties like their surface area, reactivity, coating, charge, and dissolution (as seen in most of previous BBN models for nanomaterial exposure assessment, for instance Money et al. 2012; Marvin et al. 2017), we based the model on the material-related properties of the composites in which they are normally used. The other set of parameters of influence is related to the shredding process. The model uses these input parameters to forecast the nanomaterial release in terms of discrete distributions of number, size, mass, and composition of released particles. We only consider the initial release of the nanomaterial, i.e., dissociation of nanomaterial from nanocomposites (Froggett et al. 2014) at the source (process) due to the time constraints in modeling nanomaterials transport and their simultaneous transformation from source to receptor or exposure (Ding et al. 2017a; Goswami et al. 2017). This is, however, one of the major perspectives for future work and should be dealt with later.

Before we present the methods and results, we would like to stress that the BBN model for nanomaterial release presented here should be seen as the base for further model development and we do not pretend to present an almost final model. The development of the BBN model should also be seen as an example to bring together expert knowledge for cases where data on the behavior of nanomaterials in (complex) systems is limited and to formalize the existing knowledge into a formal model.

Methods

Bayesian belief network theory

As described earlier by Marcot (2012) and Marvin et al. (2017), BBN is a probabilistic graphical model that represents probabilistic relationships among a set of nodes via a directed acyclic graph. The nodes, in turn, represent random variables $U = \{A_i, A_j, \dots, A_n\}$ with a respective set of mutually exclusive states. The directed links, i.e., the arrows between the nodes, indicate the relationship among them. A node A_i is the parent of the child node A_j , if there is a link from A_i to A_j . BBN specifies a unique joint probability distribution of all nodes given by the product of all conditional probability tables specified in BBN:

$$P(U) = \prod_{i=1}^n P(A_i | pa(A_i)) \quad (1)$$

In Eq. 1, $pa(A_i)$ are parents of node A_i and $P(A_i | pa(A_i))$ specifies a conditional probability distribution. The calculations are based on Bayesian theory, where the probability of event A at the condition of event B is expressed as:

$$P(A|B) = \frac{P(B|A) \times P(A)}{P(B)} \quad (2)$$

In Eq. 2, $P(A)$ is the a priori probability of A , $P(B|A)$ is the *conditional probability* of B under the condition of a known event A , and $P(B)$ is a priori probability of B .

Process of BBN model development

The development of a BBN heavily leans on the input from experts. As there is not enough data available in

the context of nanomaterials release, their knowledge is crucial in deciding the variables affecting the release and their dependence in the form of conditional probabilities. A similar approach has also been used in the past to generate the conditional probabilities (Money et al. 2012; Marvin et al. 2017). From the start of the model development, 10 experts with different expertise and professional backgrounds like aerosol science, material science, epidemiology, and statistics were involved. The experts were selected from within and outside the FutureNanoNeeds consortium. Experts from the shredder manufacturing and resale and from the recycling industry were interviewed to gather information on shredder configuration and use. The experts involved could be characterized mainly as generalists and subject matter experts (Knol et al. 2010). The process of the development was led by a BBN expert. Although expert selection criteria were not made formally, three selection criteria were used in selecting, especially the external experts. These criteria were:

- Expert knowledge of ENM release to air
- Expert knowledge of particulate matter release from physical processes especially shredding
- Expert knowledge on physical material properties affecting particulate matter release to air

More details on the experts involved are provided in the [Supplementary Information](#). Release data of nanoparticles were collected from 25 studies reported in the scientific literature in the period of 2007–2015. In addition, information was also used from commercial shredder manufacturers. This information was retrieved by contacting manufacturers directly and inquiring for the knowledge needed to detail-specific shredder-related parts of the model. A five-step structured approach was followed, based on the general knowledge acquisition process, to develop the graphical structure and to elicit the needed prior probabilities. The five steps taken were:

1. Identification of the most important variables (both material and shredding process related)
2. Identification of the conditional links between these variables
3. Determination of the possible states for each variable
4. Review, refinement, and completion of the graphical structure (variables and dependencies)

5. Determination of the conditional probabilities for dependencies between variables, in so-called conditional probability tables (CPTs)

The steps 1–4 were based on a combination of structured brainstorming, individual inputs, and group discussions/reviews. In the steps 1 and 2, the experts attempted to develop a structured model in which the main parameters and their relationships were included that predict the release of nanomaterials during shredding of waste. In this way, the scientific understanding was formalized to a certain extent. This approach is useful if case-measured data are lacking, see, e.g., (Kandlikar et al. 2007). It was an iterative process, with each iteration improving the understanding of the subject matter by thinking about how this should/could be modeled in a BBN, which factors are indeed relevant, how are they defined, and how do they depend on the other relevant factors. Going through the iterative process, an increased group understanding among the experts was achieved which indeed benefitted the (quality of the) model. For step 5, the a priori probabilities were determined for each variable individually using an expert elicitation process based on a Delphi-like approach. This approach ensured that different opinions of the experts are all taken into account, while offering an open podium to share arguments for certain assessments. The approach works in the following way:

1. Determine which experts will join the assessment for the variable at hand as not every expert had relevant expertise to give an assessment for each variable (list of experts in the [Supplementary Information](#)).
2. Determine if these experts together (as group) have enough expertise to assess the a priori and conditional probabilities. If not, more expertise or specific information was obtained (e.g., from shredding manufacturers).
3. Perform the elicitation process:
 - a. Each expert notes down his individual assessment for each probability in the CPT of the variable at hand
 - b. Collect the individual assessments and determine the average
 - c. Share and discuss the assumptions/underlying understanding each of the experts used when determining their individual assessments

- d. Show the assessments, both individual and average, and discuss the differences between them
- e. Give option to each expert to revise their individual assessment in light of the discussion/arguments of other experts
- f. Consensus on the average values

As a result, a total of 1089 probabilities were elicited to build 22 CPTs—1 CPT per variable. The CPTs are shown in the [Supplementary Information](#). Eventually, consensus was reached for all probabilities (all experts agreed on the use of the average, after one or more revision rounds). All structures and CPTs were implemented using GeNIe v2.0.4779.0 (Decision Systems Laboratory; University of Pittsburgh).

The model

Figure 1 shows the BBN model. It consists of four types of variables: (i) input variables which account for the material related properties/parameters (shown in blue color; e.g., hardness composite, nano-object size, brittleness, composite) and shredding process-related properties (shown in green color; e.g., number of shafts, teeth, width of knives), (ii) Switch variable (shown in orange

color, i.e., choice between primary or secondary shredder) which accounts for the adjustment needed between two possible uses of the model, (iii) intermediate variables (shown in white color; e.g., comminution potential, net energy, generated surface) which combine the effect of input variables together, and (iv) goal variables (shown in yellow color; e.g., number, size, composition, and mass of released particles) which characterize the release of particles during shredding. The combination of these variables is therefore predictive for release.

In Tables 1 and 2, material and process-related input variables are enlisted respectively with their description, the states in which they are considered to vary and their a priori probabilities elicited by the experts within the scope of selected VCs. The description and elicited a priori probabilities for the intermediate variables are provided in the [Supplementary Information](#). The whole model can be accessed at <https://www.futurenanoneeds.eu/outputs/fnn-bbn-shredding-model/>.

Sensitivity analysis

To investigate the sensitivity of the output variables, i.e., number, size, mass, and composition of released particles with regard to the input parameters (including both

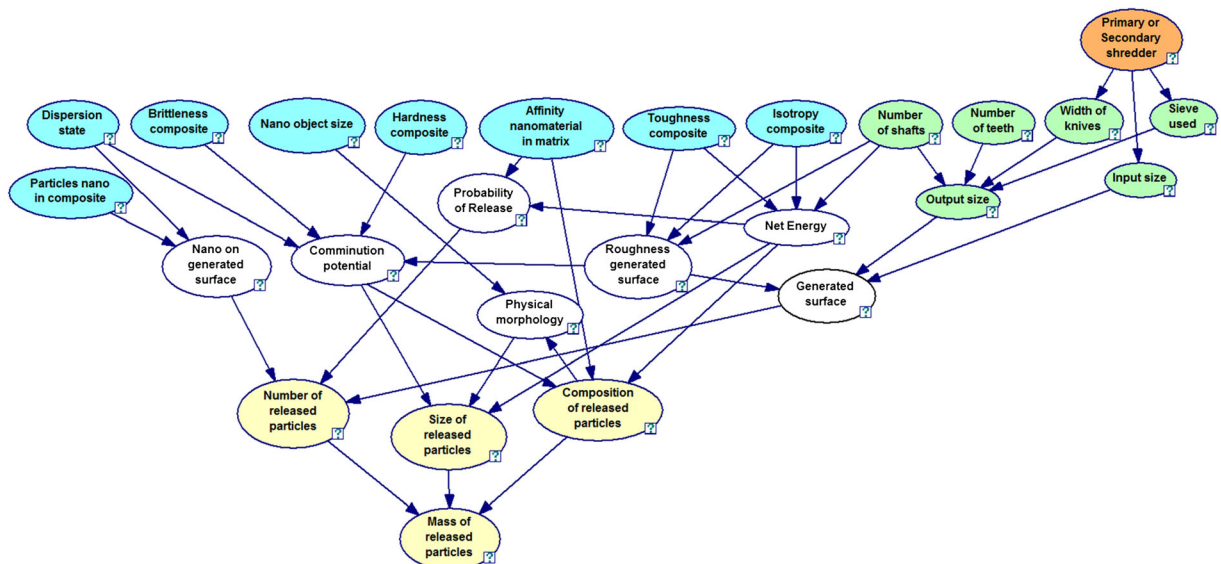


Fig. 1 Graphical structure of the BBN model. The material-related input parameters are in blue-colored nodes; the process-related input parameters are in green-colored nodes; a process

switch variable is in orange-colored node; the intermediate variables are in white-colored nodes; and the goal variables are in yellow-colored nodes

Table 1 Material related input variables of the BBN model

Variable	Description	Variable states	A priori probability in each state
Hardness composite (Mohs scale)	This variable characterizes the scratch resistance of different materials to shred through the ability of a harder material to scratch a softer material. To shred a composite, it should be less hard than the shredder blades which are generally made of hardened steel and have hardness values between 7 and 8 on the Mohs scale.	State 1: 3–4 State 2: 5–6 State 3: 7–8	State 1: 33% State 2: 37% State 3: 30%
Nano-object size (nm)	It is the primary particles size of the nanomaterial (< 100 nm, in isolated or aggregated or agglomerated form) with which the composite is reinforced.	State 1: < 10 State 2: 10–50 State 3: 50–100	State 1: 10% State 2: 50% State 3: 40%
Isotropy composite	This variable indicates the degree of uniformity towards the mechanical stress, applied via shredding, inside the material to shred.	State 1: Isotropic State 2: Intermediate State 3: Anisotropic	State 1: 17% State 2: 48% State 3: 35%
Toughness composite	A certain amount of energy is required to break down the material during shredding. This energy is indicated in terms of its “Toughness.” A tough material tends to absorb a lot of energy and deform rather than break/fracture (e.g., metals) while a not tough material tends to fracture as soon as certain energy is applied (e.g., chalk). Mathematically, toughness of a composite can be defined as the amount of energy absorbed per unit volume prior to fracture.	State 1: Tough State 2: Not tough	State 1: 70% State 2: 30%
Brittleness composite	A ductile and brittle material breaks down with and without significant deformation respectively.	State 1: Brittle State 2: Ductile	State 1: 52% State 2: 48%
Nanoparticles in composite (%)	It accounts for the mass of the nanomaterial per unit mass of the matrix expressed in %.	State 1: < 1 State 2: 1–10 State 3: > 10	State 1: 20% State 2: 60% State 3: 20%
Affinity nanomaterial in matrix	Based on prior chemical treatments or no treatments, the adhesion of the nanomaterial to the matrix in a composite may considerably vary. Between the two components, there can be (i) no bonding due to almost no adhesion (as in mere physical enclosure of nanomaterial inside the matrix), (ii) weak bonding like van der Waals forces, (iii) strong bonding via morphological structures, or (iv) chemical bonding with bonding strength similar to the matrix material itself. In the first three states, it is assumed that the bonding between the two components has a failure strength less than that of the matrix itself. For the fourth state, it is otherwise, i.e., the composite tends to break in the matrix rather than at the interface when shredded.	State 1: Not bonded State 2: Weakly bonded State 3: Strongly bonded State 4: Chemically completely inbound	State 1: 9% State 2: 29% State 3: 53% State 4: 9%
Dispersion state	It is an indicator of how well the nanomaterial is spread inside the matrix. The nanomaterial can be (i) well dispersed, i.e., each nanomaterial in full contact with the	State 1: Well dispersed State 2: Moderately dispersed	State 1: 25% State 2: 19%

Table 1 (continued)

Variable	Description	Variable states	A priori probability in each state
	matrix, or (ii) moderately dispersed, i.e., some loose nanomaterial agglomerates or their concentration gradients or both, or (iii) fully agglomerated, i.e., presence of lumps of nanomaterial which are partially or not at all in contact with the matrix, or (iv) present as a thick layer on the matrix (e.g., coating on the original product or a sandwiched structure of the original product). In the present model, this variable also accounts for the distribution of the energy absorbed by the composite before it breaks during shredding.	State 3: Fully agglomerated State 4: Layered	State 3: 7% State 4: 49%

material and process parameters), sensitivity analysis (SA) of the proposed BBN model was done using a one-at-a-time (OAT) method. This also helped to identify those input parameters where new information

would lead to the greatest reduction in uncertainty in the forecasts and identify knowledge gaps. It is important to note that OAT methods cannot account for the interaction among different input parameters. All

Table 2 Process related input variables of the BBN model

Variable	Description	Variable states	A priori probability in each state
Primary or secondary shredder	This variable is a switch to choose whether the material is being shredded for the first time (primary) or for the second time (second time). Since the choice entirely depends on the user, therefore, no variable states and, hence, a priori probabilities can be elicited for it.	–	–
Number of shafts	A shredder unit is characterized by a single or multiple counterrotating shafts (turning towards each other). A single shaft configuration offers an advantage of stringent output particle size control and a material breaking action similar to grating. Because the system operates at higher rotor speeds (80–120 rpm, sometimes even higher), frequent maintenance and part replacement are likely to occur should the cutter encounter some heavy metals. On the other hand, a multiple shafts configuration operates at lower speeds (10–30 rpm) with a material breaking action similar to cutting. It is largely used because of its viability to process heavy metals and low damages to the equipment. In the present model, this variable accounts for the total energy provided by the shredding system to break the material.	State 1: Single State 2: Multiple	State 1: 5% State 2: 95%

Table 2 (continued)

Variable	Description	Variable states	A priori probability in each state
Width of knives (cm)	The shaft(s) in a shredder are keyed to a series of knives/cutter discs. Depending on the type of application and the material to shred, the width of knives varies over a large range, generally from 1 to 5 cm for the shredding of plastic substrates, including WEEE. However, for certain applications where heavy metals and heavy power requirements are involved, width of knives is generally > 5 cm. With the increase in the width of knives, the output size decreases.	State 1: < 2 State 2: 2–5 State 3: > 5	State 1: 3% State 2: 73% State 3: 25%
Number of teeth	The number of teeth per knife generally varies from 1 to 7, depending on the material to shred and to which extent. There are some cases where the number of teeth can be greater than 8 when the required output shredded size is much smaller than in normal cases.	State 1: No teeth State 2: 1–7 State 3: 8–15	State 1: 0% State 2: 90% State 3: 10%
Sieve used	Use of a sieve or screen during shredding is decided on the need of a subsequent reduction of the output particles size in a closed loop. The output particles (once shredded) can be fed back into the shredder for further shredding if they are coarser than the sieve size, thus allowing to have a uniformly controlled maximum output particles size. Hence, there may or may not be a sieve present in a shredder depending on the shredding output requirement.	State 1: Yes State 2: No	State 1: 38% State 2: 62%
Input size (cm ²)	It accounts for the physical dimensions of the materials which are fed into a shredder.	State 1: < 1 × 1 State 2: 1 × 1–5 × 5 State 3: 5 × 5–10 × 10 State 4: > 10 × 10	State 1: 7% State 2: 29% State 3: 20% State 4: 44%
Output size (cm ²)	It considers the final physical dimensions of the shredded material as specified by the manufacturer.	State 1: < 1 × 1 State 2: 1 × 1–2 × 2 State 3: 2 × 2–5 × 5 State 4: > 5 × 5	State 1: 28% State 2: 25% State 3: 35% State 4: 12%

options of the input parameters are therefore considered equally likely. Although interaction of parameters may occur in actual release scenarios, the large number of theoretical combinations ($> 10^3$) of input parameter prohibits an attempt to systematically account for all potential interactions and most of the time they are unknown. It is reasonable to assume that the uncertainty in the SA introduced by not accounting for interactions is much smaller compared with using unknown probability distributions of the parameter options (Riedmann et al. 2015). To carry out the analysis, the base value is taken

as the arithmetic mean of the output variables when all the input variables are in their respective state 2. The input variables were then varied to either extremities, i.e., towards states 1 and 3 (only towards state 1 for the variables with two states) to compute its effect on the particular output variable. While doing this, only one input variable was varied at a time and the others were kept fixed. To vary the state of an input variable, its a priori probability was put to 100% first for state 1 and then to state 3. The complete calculations are shown in the [Supplementary Information](#).

Results

Model results

Table 3 shows the information on the output variables, i.e., *number, composition, size, and mass of released particles* and their distributions as predicted by the model. These results are produced by feeding the model with a priori probabilities shown in Tables 1 and 2 for the input variables pertaining to PV panels, Li-ion batteries, and electronic displays.

Prediction capacity of the model

To evaluate the capacity of the BBN model in predicting initial nanomaterial release during shredding, its predicted results need to be compared with experimental cases which together cover the possible situations (input) one might encounter during shredding. Individually, each case is run in the model, after which the outcome of the model in each case is compared with the experimentally determined output data from the respective case.

However, only one experimental case (Raynor et al. 2012) was found in the literature that deals with the measurement of particle release around the shredder. This experimental case is referred to as test case 1 in the present study. It involved the shredding of a commercial product—18CPP091 Forte Nanocomposite—manufactured by Noble Polymers (Grand Rapids, Michigan, USA) which was composed of polypropylene resin reinforced with approximately 5%, by mass of montmorillonite nanoclay. The shredding was performed using a 4-kW small-scale industrial grade shredder (Cumberland Engineering Corp., South Attleboro, MA). The released particle concentration measurements were taken adjacent to the shredder, i.e., with negligible distance between source and measurement probe. It must be noted that the addition of nanoclay reduced the number concentration found by FMPS during shredding.

There is another shredding related laboratory test whose results are not yet available in the literature. It was carried out at CEA-LITEN-NanoSafety Platform-Grenoble, France. It concerns the shredding of nano-enabled electrodes (cathode) made of a mixture of

Table 3 Goal variables of the BBN model

Variable	Description	Parent nodes	Variable states	Conditional probability in each state
Number of released particles (in g ⁻¹)	It accounts for the number of particles released/aerosolized during shredding of a unit gram of the material prior to any transformation.	1.1.1.1. Nano on generated surface 2. Probability of release 3. Generated surface	State 1: < 10 ³ State 2: 10 ³ –10 ⁶ State 3: 10 ⁶ –10 ⁹ State 4: > 10 ⁹	State 1: 64% State 2: 23% State 3: 7% State 4: 6%
Composition of released particles	Based on the parent nodes, the released particles may vary in their composition.	1.1.1. Net energy 2. Affinity of nanomaterial in matrix 3. Dispersion in matrix	State 1: Individual pure nanomaterial State 2: Agglomerated or aggregated pure nanomaterial State 3: Composite of both nanomaterial and matrix	State 1: 15% State 2: 42% State 3: 43%
Size of released particles (in nm)	It accounts for the initial size of the released particles prior to any transformation.	1.1.1. Net energy 2. Physical morphology 3. Comminution potential	State 1: < 10 ² State 2: 10 ² –2.5 × 10 ³ State 3: 2.5 × 10 ³ –10 ⁴	State 1: 27% State 2: 43% State 3: 30%
Mass of released particles (μg/kg)	For a given mass of the material to shred, it accounts for the total mass of the released particles.	1.1.1.1. Number of released particles 2. Size of released particles 3. Composition of released particles	State 1: < 10 ¹ State 2: 10 ¹ –10 ³ State 3: 10 ³ –10 ⁶ State 4: > 10 ⁶	State 1: 76% State 2: 13% State 3: 4% State 4: 7%

LiFePO₄ mixture (LFP), Vapor-Grown Carbon Fiber (VGCF), conductive carbon black (Super P, Super C65) deposited on an aluminum foil. It is referred to as test case 2 here. For this study, a 3-kW high-torque shredder (SM300 cutting mill, Retsch GmbH, Haan, Germany) equipped with a bottom sieve (5 × 5 mm²) was operated at 1500 rpm. All direct-reading instruments were connected to the same sampling probe that was positioned approximately at 30 cm from the top of the shredder which represents the breathing zone for the workers feeding the shredder. Clearly, the experimental results, obtained during this test case, are not on the *initial release* as defined within the scope of the model and its use (for industrial scale shredders) but related to the occupational exposure assessment. The data available for the two test cases is shown in Table 4.

To obtain the model predicted results corresponding to the two test cases, the a priori probabilities were set to 100% for the states shown in Table 4 (i.e., 100% probability for state 2 in case of *hardness composite*; state 1 in case of *nano-object size*, etc.). The obtained

probabilities for four output variables were noted and are shown in Table 5. For test case 1, all four goal variables have an agreement between their model predictions and experimental values. The model showed 79% of the released particles to be of a composite nature, of which some can be matrix material only. The current model does not have the ability to predict the release of matrix material only. For the test case 2, the state 2 is most probable for both *composition of released particles* and *size of particles* as per the model which also agrees with the experimental data from the case too. However, *number of released particles* is underestimated by the model (i.e., state 1) as the experimental value is higher than predicted. The experimental values for the mass of released particles were not available for this test case. If the model predicted values are split almost equally among two states (e.g., states 1 and 2 for size of released particles in both test cases), it can be interpreted as released particles have two size modes. In such a case, although partial, we mention an agreement between predicted

Table 4 Values of the input variable for two test cases

Variables	Test case 1 Variable state	Test case 2 Variable state	Remarks (if any)
Hardness composite	State 2: 5–6	State 1: 3–4	
Nano-object size	State 1: < 10 nm	State 3: 50–100 nm	
Isotropy composite	State 1: Isotropic	State 3: Anisotropic	
Toughness composite	State 1: Tough	State 2: Not tough	
Brittleness composite	State 2: Ductile	State 2: Ductile	For test case 2, a clear distinction is hard to make as the test sample is ductile in bulk but when unrolled, the deposit is moderately brittle (i.e., releases dust when crumpled)
Particles nano in composite	State 2: 1–10%	State 2: 1–10%	Due to its confidential nature, the manufacturer did not disclose the exact formulation of the electrode coatings but the range is correct.
Affinity nanomaterial in matrix	State 4: Chemically completely inbound	State 2: Weakly bonded	
Dispersion state	State 1: Well dispersed	State 4: Layered with metallic foil	
Primary or secondary shredder	Primary	Primary	
Number of shafts	State 1: Single	State 1: Single	
Width of knives	State 2: 2–5 cm	State 3: > 5 cm	
Number of teeth	State 2: 1–7	State 1: No teeth	
Sieve used	State 1: Yes	State 1: Yes	
Input size	State 4: > 10 × 10 cm ²	State 4: > 10 × 10 cm ²	
Output size	State 1: < 1 × 1 cm ²	State 1: < 1 × 1 cm ²	

Table 5 Comparison between experimental and model predicted results to determine the predictive capacity of the model

Goal Variable	Test case 1			Agreement: Exp. and model predicted result	Test case 2			
	Experimental result	Model predicted result			Experimental result	Model predicted result	Agreement: Exp. and model predicted result	
Number of released particles	The total number concentration (shown by FMPS) produced by a composite plaque (mass = 28.4 g) = 26000 cm ⁻³ (after removing background particles).	State 1: <10 ⁷ g ⁻¹ State 1: 73 State 2: 22 State 3: 5 State 4: 0		Yes	The total number concentration (shown by FIDAS) produced by 3 kg electrode = 1860 cm ³ State 2: 10 ³ -10 ⁶ g ⁻¹	State 1: 56 State 2: 30 State 3: 12 State 4: 2		No
Composition of released particles	Scanning electron microscopy show that no nano-clay particles separated from the matrix.	State 3: Composite State 1: 8 State 2: 13 State 3: 79		Yes	Scanning electron microscopy show micrometric agglomerates of pure nanomaterial carbon State 2: Agglomerated or aggregated	State 1: 21 State 2: 58 State 3: 21		Yes
Size of released particles	The size mode measured by FMPS= 11 nm.	State 1: <10 ² nm State 1: 49 State 2: 41 State 3: 10		Yes	The size mode measured by FIDAS= 250 nm State 2: 10 ² -2.5 x 10 ³ nm	State 1: 37 State 2: 44 State 3: 19		Yes
Mass of released particles	For a plaque getting shredded in 15 s, the mass of particles measured by DustTrak= 0.014 mg.m ⁻³ in an air flow rate of 323 m ³ .h ⁻¹ .	State 1: <10 µg/kg State 1: 88 State 2: 10 State 3: 2 State 4: 0		Yes	N.A.	N.A.	-	N.A.

Further instrumental details are shown in the [Supplementary Information](#)

and experimentally observed values. Even though the outcomes seem overall promising, nevertheless, no final conclusion can be drawn here on the predictive capacity of the model on nanomaterial release—neither in the favor of the model (as only one test case to support) or against (as experimental conditions of test case 2 are out of the scope of the model). Moreover, both test cases are fairly similar in input values to some extent, and therefore, they do not test the parameter space of the model. The availability of more experimental data is crucial to reach any conclusion.

Sensitivity analysis

In Fig. 2, we show the tornado diagrams. Highly sensitive input variables affect the results more significantly and appear towards the top of the diagram. Corresponding to each input variable, its spread is also shown by taking the absolute of the difference between two extreme points of the corresponding bar. The spread allows the quantification of the sensitivity.

Figure 2a shows that *number of released particles* is most sensitive to *input size* with a negative correlation, i.e., by increasing the input size of the material to shred (input size state 1 → state 3), the number of released

particles decreases. For the second most sensitive parameter, i.e., *output size* the sensitivity decreases by tenfold ($6 \times 10^{10} \rightarrow 6 \times 10^9$). The subsequent less sensitive input variables in the order of decreasing sensitivity are (i) *isotropy composite* (i.e., anisotropy induces an increase in the number of released particles), (ii) *sieve used* (i.e., use of a sieve increases the number of released particles), (iii) *primary or secondary shredder* (i.e., shredding the already shredded material increases the number of released particles), (iv) *width of knives* (i.e., increasing the width of knives increases the number of released particles), and (v) *particles nano in composite* (i.e. increasing the mass% of the nanomaterial in per unit mass of the matrix increases the number of released particles). *Mass of released particles* also has similar results of sensitivity analysis (see Fig. 2b) which is understandable from the fact that it has *number of released particles* as one of its parent nodes.

Number of shafts which signifies the rotating speed of the shredder affects the *size of released particles* most, as seen in Fig. 2c. A configuration transition from single shaft (operating at higher speed of 80–12 rpm) to the multiple shafts (operating at lower speed of 10–30 rpm) increases the size of the released particles. While the increase in the *affinity of nanomaterial in*

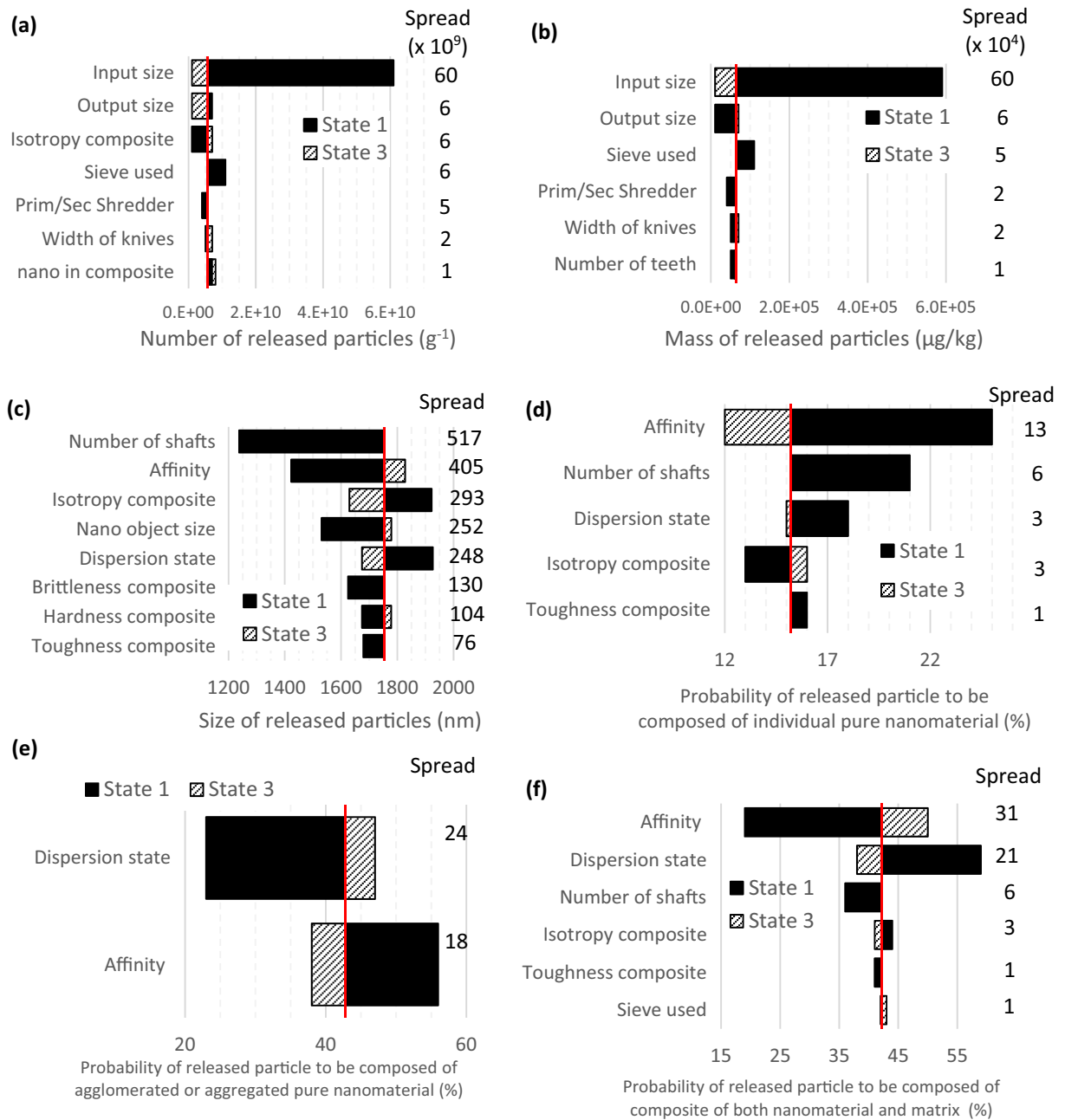


Fig. 2 Sensitivity analysis of the model showing the most influential input variables and the magnitude of their influence (in terms of bar spread) for **a** number, **b** mass, **c** size, and **d–f** composition of released particles

matrix, isotropy composite, nano-object size, and hardness composite increases their size, an increase in the dispersion, brittleness composite, and toughness composite decreases their size.

For the composition of released particles, affinity nanomaterial in matrix and their dispersion state are the most effective input variables. The strong bonding of

the nanomaterial to the matrix increases the probability of the nanomaterial to release in association with the matrix, i.e., in the composite form. Consequently, the release of nanomaterial, either individually or as agglomerated/aggregated, becomes less probable. This can be easily observed in Fig. 2d–f. Similarly, if the nanomaterial is not well dispersed in the matrix, the

probability of its release in an agglomerated form is higher than as individual or composite. The only process parameter which is relevant in deciding the composition of the released particles is the *number of shafts*. A single shaft system tends to release individual nanoparticles due to higher input energy.

Discussion

Various case studies which were carried out within the framework of the FutureNanoNeeds project and focussed on the value chains pertaining to the use or treatment of different nanomaterials concluded that in all instances the end-of-life stage is critical with respect to emission to environmental compartments and potential for transformation. It appeared that the potential release of nanomaterial containing e-waste during recycling processes (e.g., shredding) and their system behavior is relatively unexplored and/or unknown. For this reason, a BBN model was developed to forecast nanomaterial release during the shredding of WEEE by considering three selected value chains case studies—PV panels, Li-ion batteries, and electronic displays. The selection of the input material variables, which were chosen to be specific to final product and not to nanomaterial, allows the model to be applicable to the shredding of a wide range of products and not limited to WEEE. The model can itself allow the development of safer-by-design (Goswami et al. 2017; Shandilya et al. 2015a) approach by testing several formulations and end-of-life scenarios without experiments.

The input variables of the model are physical endpoints which are considered to be sufficient and necessary to predict release. All other properties which seem to be relevant can either be described by these input variables or have these input variables as endpoints. For example, for the type of used polymer or surface coating of nanomaterial or presence of additives (dispersing agents, etc.), the variable *affinity nanomaterial in matrix* can be an endpoint affecting release. To include shredding process parameters, a priori probability elicitation were done considering different power/energy configurations, automatic and manual feeding, different designs for different size ranges of fragments produced, different collection systems of fragments (e.g., gravity, vacuum, etc.), different sized or configuration of shredder (including throughput), open or enclosed shredder (e.g., encasings), and ventilation systems.

Limitations

Scoping of the shredding case prior to the expert meetings was crucial in order to narrow down the system boundaries and the developed network. The choice of the material properties, shown in Table 1, does not pertain to nanocomposites reinforced with fibrous nanomaterials like nanofibers and nanotubes, and hence, they are excluded from the model. These fibrous nanomaterials have particular physical and chemical properties due to their high aspect ratio. Consequently, their release behavior is assumed to be different from that of particulates. Additionally, and although we did not address the health aspects of the materials released, the high risk of these nanofibers was taken into consideration for excluding them. Like asbestos fibers, fibrous and low solubility nanomaterials are shown to have asbestos like properties and are able to penetrate the pleural cavity and cause inflammations there (Xu et al. 2012). If these nanomaterials were to be included, one or more separate/additional sets of input material parameters would be required which account for their specific characteristics such as heat transfer, vibrations, tensile strength, and extremely high specific surface area. These sets should be based on nanofibers having the same properties in common such as tensile strength, heat conductivity, and specific surface area. Then switch variables (asking to select either a fibrous material group or particulate nanomaterial) would be required which allows their selection (as is done in the present article for the choice between primary or secondary shredder). Inclusion of these materials into the BBN model would call for validation of the influence of their parameter sets on the predicted results.

The release is assumed to occur (if relevant) within the Local Control Influence Region (LCIR) which is a virtual boundary around a source that represents the zone of influence of any local control system (Schneider et al. 2011). The release to the environmental compartments (air, soil, and water) and waste flow processes (wastewater treatment plants, waste-in-storage, landfills) are excluded from the scope of the present model. The “wet” shredding process which uses coolant is also excluded. The time scale is not considered in the current release scenario, since it would add much complications to the calculation (feeding rate, release rate, etc.). The advantage of such an approach is to have a simplified yet effective starting point (baseline) which can be later extended to nanomaterial flows to waste streams.

Predictive capacity of the model

Although a clear agreement was observed between the experimental and model predicted results for test case 1, a single test case does not confirm the prediction power of the model. The conditional probabilities for the developed BBN model are assessed by experts in different domains. The obtained assessments might be inaccurate and over- or underestimated as a consequence of incompleteness of data and partial knowledge of the domain. Since the output probabilities of a network are built from these assessments, they may be sensitive to the inaccuracies involved and may even be unreliable. This may lead to a consistent discrepancy in the model output and the experimental results, once they are available. This discrepancy can be eliminated by using the sensitivity analysis outcome of the model (Section 3.3) and by identifying which model variables can be modified to arrive at the expected output. This can be achieved by taking one of the individual opinions (implying a different distribution of a priori probabilities) instead of the group consensus. Such a model variable modification may also require gaining further knowledge by involving additional experts on a specific subdomain. A different combination of expert opinions would lead to an improved output (if differences can be narrowed down to originate at a specific variable). For example, if some experts take a certain effect into account and other experts do not, what would be the resulting CPT if only the experts with (or without) taking this effect into account are combined? Does this CPT lead to an improved output? For now, there is no way of gaining further significant knowledge unless more pertinent experimental data becomes available.

The beliefs of the experts which were used during the model development stemmed from the observations during prior exposure related studies which tested the effect of mechanical or environmental solicitations of nano-enabled products (Hsu and Chein 2007; Li et al. 2008; Reijnders 2009; Gohler et al. 2010; Wohlleben et al. 2011; Sachse et al. 2012; Shandilya et al. 2014b, 2015b; Ding et al. 2017b). When the sensitivity analysis outcome is seen in the context of these studies, we find those beliefs to be still valid and consistent. For example, when polymer nanocomposites/paints are subjected to UV irradiation (Hsu and Chein 2007; Li et al. 2008; Reijnders 2009; Shandilya et al. 2015b; Wohlleben et al. 2011), an evaluation (direct or indirect) of the effect of the affinity of the nanomaterial to the matrix on the

nanomaterial release can be done. Such an irradiation leads to an inevitable degradation of the polymer matrix and further increase in its brittleness, which consequently reduces the adhesion strength between the nanomaterial and matrix. An increase in the number of released particles and release of pure nanomaterial (individual or agglomerate) are typical observations in these types of studies. Moreover, studies employing the use of different mechanical solicitation means, like slight or severe abrasion (Taber abrasion, manual or machine sanding), drilling, and cutting (Ding et al. 2017a; Gohler et al. 2010; Sachse et al. 2012; Shandilya et al. 2014b), conclude in general that higher magnitude of input mechanical energy leads to the reduction in the released particle size mode.

Conclusions and recommendations

The present BBN model estimates the nanomaterial release by characterizing it in terms of discrete distributions of four goal variables—number, composition, size, and mass of released particles. The fibrous structures with high aspect ratio are kept excluded from the scope of the model at current stage. Fifteen system components (eight material and seven process properties) were used to identify relevant release parameters for shredding. A combination of these components is applied in the BBN to predict release, with a causal relationship between the underlying variables of each component. The total number of probabilities to elicit was over a thousand and required the consensus of all experts. The developed BBN shredding model can be used for a wide range of nanocomposites and adapted for comminution activities like milling or crushing. The model output of release can also provide input or eventually be linked with dispersion and exposure models.

For the shredding of PV panels, Li-ion batteries, and electronic displays, the model currently suggests that there is a 64% probability that the released particles will have a number less than 10^3 particles/g of the shredded material (or mass less than $10 \mu\text{g}/\text{kg}$ of the shredded material) with 43% probability of them having size between 100 and 2500 nm. There is an almost equal probability of them getting released in the form of agglomerated or aggregated pure nanomaterial (42%) and composite of both nanomaterial and matrix (43%). The model was also analyzed by evaluating its

output vis-à-vis two specific experimental test cases. The experimental results from the test cases were found to be in general agreement with the model predicted results. However, the data is too limited to draw final conclusions at this stage. The sensitivity analysis indicated that the number (and hence the mass) of the released particles are primarily controlled by the size of the material fed to the shredder (i.e., *input size*). In contrast, controlling the size of the released particles is rather complicated as it is controlled by various variables like affinity between nanomaterial and matrix, isotropy of the composite, nano-object size, and the amount of the energy input (represented by *number of shafts*). *Dispersion state*, *affinity*, and *number of shafts* are the most influential ones to decide the composition of the released particles.

With the availability of more field data and consequently with more specific sensitivity analyses, these outcomes could provide some guidance to further investigate the reliability of the model output with respect to the conditional probabilities and to shed light on the significance of input parameters in relation to output results. To enhance the quality of the prediction of release of nanomaterials from shredding nanomaterial containing WEEE, it is recommended to select the experts using criteria (see, e.g., Hung et al. 2008; Kandlikar et al. 2007; Knol et al. 2010; Roman et al. 2012) like substantial contribution to scientific literature in fields like nanomaterial and/or particulate matter release due to physical processes, the capacity and willingness to contribute to further development of the BBN model; and openness to accepting deviating opinions. In case the model is extended to predict exposure of workers, which it does not now, the set of process-related input variables has to be extended with parameters that describe local exhaust and air filtration systems and variables that further describe the local control influence region (Tielemans et al. 2008). The prediction of the worker exposure in the future model should also include the fate of initially released nanomaterials when entering the worker's near field.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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