Journal of Hazardous Materials 192 (2011) 837–845

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Assessing the environmental impact of five Pd-based catalytic technologies in removing of nitrates

Pei-Te Chiueh*, Yu-Hung Lee, Chih-Yi Su, Shang-Lien Lo

Graduate Institute of Environmental Engineering, National Taiwan University, 71 Chou-Shan Rd., Taipei 106, Taiwan

ARTICLE INFO

Article history:
Received 17 February 2011
Received in revised form 29 May 2011
Accepted 30 May 2011
Available online 6 June 2011

Keywords:
Life cycle assessment
Analytic hierarchy process
Environmentally friendly
Nitrate
Catalytic denitrification

ABSTRACT

Emerging technologies involving chemical catalytic processes to remove nitrate from water have proven efficient and cost-effective. However, the environmental impact of noble metals and metals at the nanoscale used in these processes has become a topic of serious concern. The aim of this research was to develop a system for evaluating the environmental impact of technologies associated with Pd-based catalytic denitrification. This research performed life cycle assessment (LCA) based on a detailed analysis of the technologies to examine the environmental burden associated with all stages of the removal process. We then applied analytical hierarchy process (AHP) to determine the weights of various burdens. We implemented the proposed system to determine the relative environmental friendliness of 5 processes used for the removal of nitrate. These five methods use Cu–Pd/TNTs, H_2 + Pd–Cu/TiO_2, Pd–Cu/TiO_2, Pd/ZnO, and Pd–Cu/FeO as catalysts for the removal of nitrate. The results indicate that the use of palladium and the consumption of electricity have a major environmental impact; while the use of Pd–Cu/TiO_2 as catalyst was the most environmentally friendly of the five processes evaluated.

1. Introduction

Nitrate pollution in groundwater originates mainly from human activity including excessive long-term use of chemical fertilizers in agriculture, intensive animal husbandry, discharge from wastewater and sewage, and leachate from landfills [1]. Excessive quantities of nitrate may enter the human body via nitrate-contaminated drinking water or foodstuffs. Once absorbed by the body, nitrate is reduced to nitrite that may combine with hemoglobin to form methemoglobin, seriously reducing the blood oxygen-carrying capacity that has been associated with ailments such as blue baby syndrome [2,3].

Current methods for removing nitrate can be grouped into two categories: (1) transport in nature such as reverse osmosis, ion exchange, and electrodialysis; and (2) destruction in nature such as biological denitrification, chemical denitrification, and catalytic denitrification [3–7]. Nitrate transported from the source requires costly follow-up treatment through further processing or disposal. Nevertheless, the efficiency of biological denitrification is influenced to a high degree by the activity of microorganisms in the water. In addition, such biological processes often require extended treatment time [7]. Chemical denitrification processes such as utilizing the reduction capability of zero-valent [8–10], bimetals [5,11], catalytic hydrogen [12], or photocatalytic reactions [13] have proved to decompose and destruct nitrate rapidly. These methods are highly efficient and may be performed at relatively low cost [14]. Nonetheless, the environmental impact of adding precious metals or nanoscale metals has become a topic of concern. Due to the recent emergence of nano-technology, information regarding the impact of this technology on the environment and human health are lacking, thereby making it difficult to manage the risk associated with utilizing, transporting, and pollution preventing used in this technology. The current strategy in dealing with the damage associated with nanoscale materials and products is the application of the concept of LCA to evaluate issues related to the environment and human health [15].

Life cycle assessment (LCA) is a systematic approach used to investigate the impact of products or technologies on the environment. Farre et al. [16] used LCA to identify the most environmentally friendly method for the removal of herbicides from water. The three methods evaluated were the artificial light photo-Fenton process, the photo-Fenton process coupled with biological treatment, and the solar driven photo-Fenton process. The results of LCA indicate that the artificial light photo-Fenton process was the least environmentally friendly, with double the environmental impact compared to the most environmentally friendly technology, the photo-Fenton process coupled with biological treatment. Regardless of the method used, the consumption of hydrogen peroxide and power are critical elements associated with the environmental impact of these methods. The assessment of environmental impact

---

* Corresponding author. Tel.: +886 2 3366 2798; fax: +886 2 2392 8830.
E-mail address: ptchueh@ntu.edu.tw (P.-T. Chiueh).

0304-3894/$ – see front matter © 2011 Elsevier B.V. All rights reserved.
doi:10.1016/j.jhazmat.2011.05.096

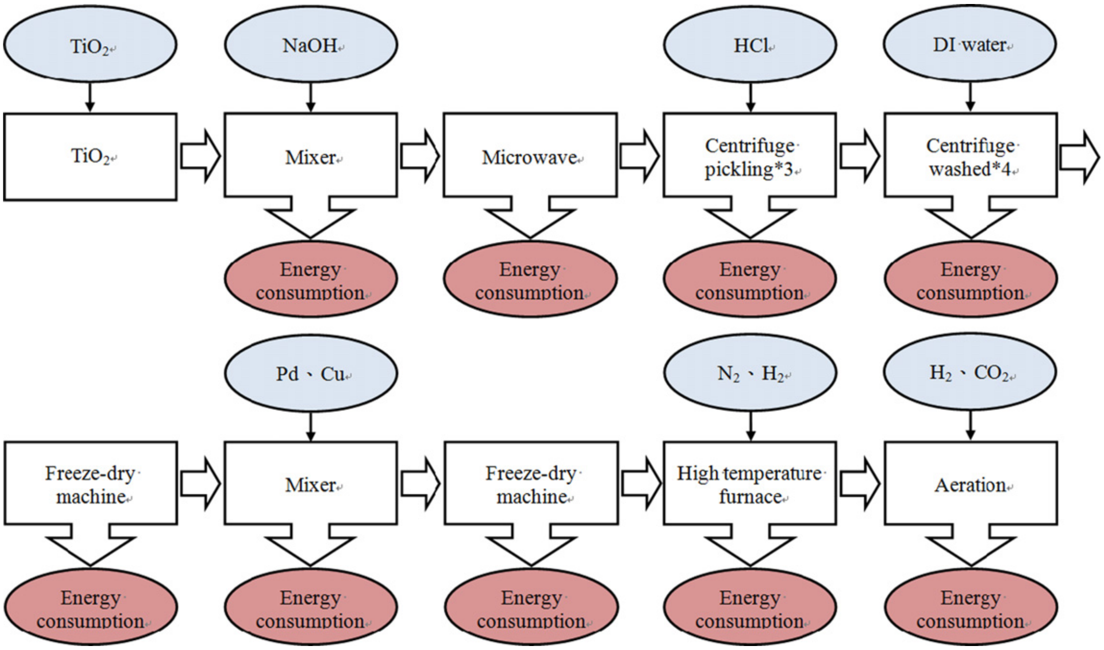

Fig. 1. Flow diagram for nitrate removal by Cu–Pd/TNTs (Case A).

in the removal of a commercial reactive dye (Cibacron Red FN-R) from wastewater using similar advanced oxidation methods was also examined by the same research group [17].

Treatment processes in environmental engineering often depend on the application of chemicals; however, the use of such chemicals may have its own impact on the environment. Shonnard et al. [18] described how BASF Chemical Company reconciled the ambivalence by developing a method for analyzing the eco-efficiency of products or processes at various stages of their life cycles to gain a better understanding of the magnitude of the environmental impact they cause, in addition to calculating the total costs of the associated processes. That method of analysis applied equal weightings to economic cost and environmental impact. If a product had low economic cost and little environmental impact, it belonged to the group of high ecological benefit, receiving a recommendation for its development.

In contrast, determining the means by which to consider multiple indices for the evaluation of environmental impact has become an important research topic. For example, Pineda-Henson et al. [19] used LCA to investigate various methods proposed to mitigate the environmental impact of the pulp and paper manufacturing process. They established 7 indices, depletion of natural resource, ecological toxicity (land and water), humantoxicity (land, air, and water), and greenhouse effect. They then employed AHP to determine the weight of each index to combine quantifying data in LCA to determine hot spots of environmental impact caused by the pulp and paper manufacturing process. The theory behind the AHP method is based on expressing the problems to be evaluated through an hierarchical structural system. The quantification of complicated and disorganized problems into simpler hierarchical structures allows decision makers to determine the priority for actions [20].

This research inventoried 5 laboratory-scale methods for removing nitrate from water, and compared the differences in environmental friendliness. These 5 methods included: (1) the Cu–Pd/TNTs method, (2) the H_2 + Pd–Cu/TiO_2 method, (3) the Pd–Cu/TiO_2 method, (4) the Pd/ZnO method, and (5) the Pd–Cu/FeO method.

2. Materials and methods

2.1. Case studies

(1) Case A: study on heterogeneous catalytic aqueous nitrate over Cu–Pd/titanate nanotubes (Cu–Pd/TNTs):

Chen [21] prepared Cu–Pd/TNTs for the catalytic removal of nitrate according to the flow diagram shown in Fig. 1. The prepared material with 20 wt% bimetal deposits was immersed in nitrate-contaminated water, bubbled with CO_2 and H_2. Under optimum treatment conditions, i.e. the pH of the solution was maintained at a constant level of pH 5, enabling the decomposition of 20 mg-N/L in 30 min. The nitrogen selectivity, which measures the portion of nitrate converted to nitrogen gas, was 71.3%.

(2) Case B: selective catalytic hydrogenation of nitrate to nitrogen using Pd–Cu/TiO_2 catalysts (H_2 + Pd–Cu/TiO_2):

Chen [22] covered titanium dioxide with bimetals of copper and palladium to form a Pd–Cu/TiO_2 catalyst, and used it to decompose nitrate in a hydrogen-enhanced system (Fig. 2). The optimum condition was reached when the solution pH was maintained at pH 7; the addition of 3 wt% with 1:1 of Pd and Cu deposits decomposed 40 mg-N/L in 2 h for a nitrogen selectivity of 73.7%.

(3) Case C: selective photocatalytic reduction of nitrate to nitrogen using a Pd–Cu/TiO_2 catalyst (Pd–Cu/TiO_2):

Tseng [23] applied the photo-deposition method to lade bimetals of copper and palladium on titanium dioxide to produce Pd–Cu/TiO_2 capable of catalytically decomposing nitrate (Fig. 3). The treatment process did not control the solution pH. Under the optimum conditions using TiO_2 deposited with 1% Pd and 1% Cu with 0.04 mL/L of formic acid as the electron captor, 40 mg-N/L was decomposed in 30 min for a nitrogen selectivity of 80%.

(4) Case D: reduction of nitrate by zero-valent Zinc and Pd/Zn bimetallic particles (Pd/ZnO):

Hong [24] laded zero-valent zinc with palladium to produce a Pd/ZnO catalyst (Fig. 4). Nitrate-contaminated water was bubbled with nitrogen gas, and maintained at pH 7 at 25 °C. The optimum condition was reached when 10% Pd laded on zero-valent zinc. The addition of 2.85 g/L of the catalyst decomposed 40 mg-N/L in 3 h for a nitrogen selectivity of 80%.

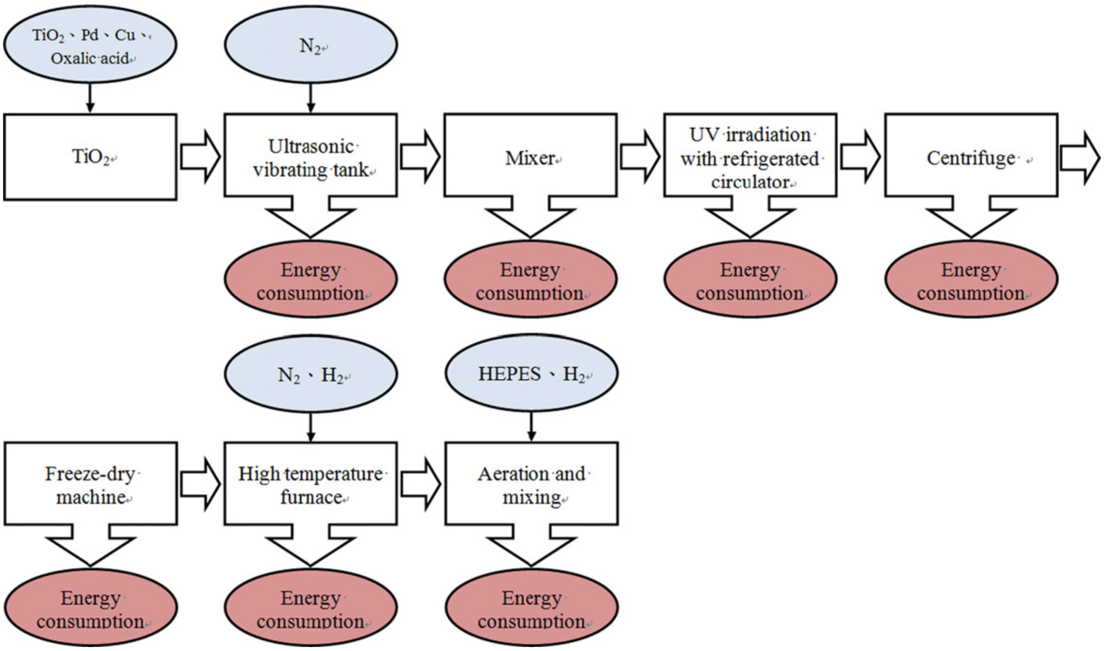

Fig. 2. Flow diagram for nitrate removal by H_2 + Pd–Cu/TiO_2 (Case B).

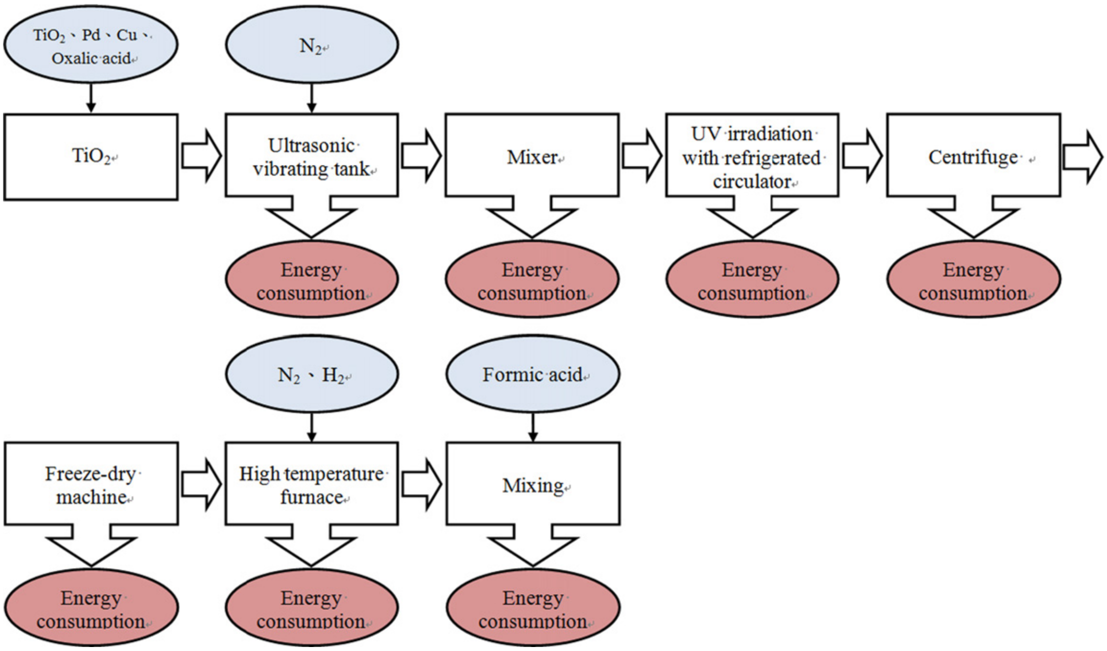

Fig. 3. Flow diagram for nitrate removal by Pd–Cu/TiO_2 (Case C).

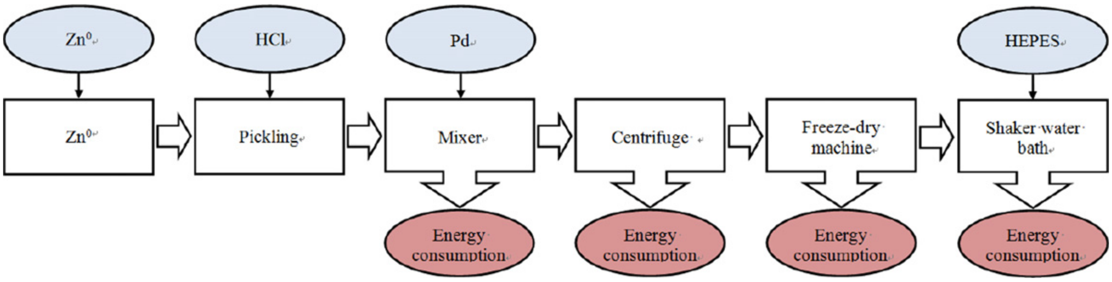

Fig. 4. Flow diagram for nitrate removal by Pd/ZnO (Case D).

Table 1
Life cycle inventory for five cases.

Case	Case A (Cu–Pd/TNTs)	Case B (H_2 + Pd–Cu/TiO_2)	Case C (Pd–Cu/TiO_2)	Case D (Pd/ZnO)	Case E (Pd–Cu/FeO)	Databases
Input						
Titanium dioxide (kg)	9.75	8.13	8.13	–	–	Ecoinvent
Sodium hydroxide (kg)	455	–	–	–	–	Ecoinvent
Hydrogen chloride (kg)	2.67	–	–	1.46	–	BUWAL 250
Copper (g)	4062.50	121.88	121.88	–	20.13	Ecoinvent
Palladium (g)	4062.50	121.88	121.88	81.25	12.19	Ecoinvent
Hydrogen (kg)	4.34	38.04	11.73	–	2.63	Ecoinvent
Nitrogen (g)	12	1625	1625	497,048	679,860.5	Ecoinvent
Carbon dioxide (kg)	28.96	35.75	14.3	60.02	60.02	Ecoinvent
Carbon monoxide (kg)	–	22.75	9.1	–	–	Ecoinvent
Formic acid (kg)	–	–	2.99	–	–	Ecoinvent
Zinc (g)	–	–	–	1625	–	Ecoinvent
Natural gas (kg)	–	–	–	19.1	19.1	BUWAL 250
Sulphuric acid (kg)	–	–	–	33.42	33.42	Ecoinvent
Iron (kg)	–	–	–	–	8.83	Ecoinvent
Electricity (kW h)	121,111	45,365	45,365	44,418	79,084	Ecoinvent
Car (km)	448.22	448.22	448.22	448.22	448.22	BUWAL 250
Output						
Atmosphere emissions						
Nitrogen (kg)	0.23	0.24	0.26	0.26	0.08	Ecoinvent
Water emissions						
Nitrite (kg)	0.09	0.09	0.07	0.07	0.25	Ecoinvent
Total costs						
Material and energy cost (NT)	8,638,763	965,439	841,313	1,959,427	1,910,038	

(5) Case E: reduction of nitrate using catalytic Cu/Pd bimetallic particles (Pd–Cu/FeO):

Tseng [25] laded zero-valent iron with bimetals of copper and palladium to produce a Pd–Cu/FeO catalyst to decompose nitrate in water (Fig. 5). The system was maintained under optimum operational conditions of 25 °C with a constant pH of 8.3. The addition of the catalyst comprising 100% iron, 0.5% copper and 0.3% palladium decomposed 40 mg-N/L in 2 h for a nitrogen selectivity of 21.8%.

2.2. *Life cycle assessment (LCA)*

In 1990, the Society of Environmental Toxicology and Chemistry (SETAC) developed LCA technology to evaluate the environmental impact of products or processes associated with the exploitation of raw materials, processing and manufacturing products, transportation, distribution, consumption and maintenance, and final waste disposal [26]. Under the ISO 14040 standard, LCA is carried out in four steps: (1) the definition of the goals and scope, (2) analysis of life cycle inventory, (3) assessment of life cycle impact, and (4) interpretation [27]. Defining the goals and scope, particularly for system boundaries and functional units, is necessary to ensure that evaluation results are consistent with the expected application. Establishing identical system boundaries and functional units provides a suitable platform with which to make meaningful comparison of the results. During the stage of life cycle inventory analysis, the entire life cycle within the defined boundaries of the inventory system, including the use of input resources, the consumption of raw materials and energy, and the waste discharged into the environment is quantified, collected, and categorized. The LCA calculations in the current study were performed using Simapro 7.1 software, according to established internal databases. There are several databases distributed along with the Simapro software, such as Ecoinvent, US LCI database, US Input Output database, Danish Input, and so on. In our study, the major data for LCA on transportation, chemicals, waste treatment and electricity, were based on the Ecoinvent 2 and BUWAL 250 databases. The electricity mix was determined by the local inventory data in Taiwan, in which coal power is the largest fraction.

Many researchers have used the Eco-indicator 99 as an impact assessment model [28–30]. It is a damage function based model, including human health, ecosystem quality, and resources as indices of the major damage categories. We selected the Eco-indicator 99 model as our LCA model in this research due to its clear corresponding significance and the ease with which it can be adapted by policy makers. We used characteristic values of 5 evaluated cases for the final stage of interpretation, to compare the extent of environmental impact and enable the proposal of meaningful, practical recommendations. These results can provide a reference for policy makers in the selection of materials or processes with low polluting potential, and low environmental impact.

2.3. *Analytic hierarchy process (AHP)*

Analytic hierarchy is generally applied to decision-making problems involving multiple criteria under uncertain conditions [31,32]. It systemizes complex problems by establishing independent hierarchies. In conjunction with weights agreed upon by experts, obtained from answers to questionnaires, AHP results enable policy makers to reduce the risk associated with decision making. Recently, AHP methods have been combined with research methods in various fields. For example, Huang and Ma [33] combined LCA and AHP for a multidimensional environmental evaluation of packaging materials; Kengpol and Brien [34] conducted research in product development based on a combination of AHP and cost benefit statistics. Details and proof of the mathematics underlying AHP can be found by Saaty [35]. This research combined the impact data obtained using LCA with the index weights of various impact factors obtained using the AHP method to evaluate the final impact grade for the 5 cases in the identification of the most environmentally friendly approach.

3. Results and discussion

3.1. *Life cycle assessment (LCA)*

3.1.1. *Definition of goals and scope*

We conducted LCA to assess 5 nitrate removal methods associated with the exploitation of materials, manufacturing of products,

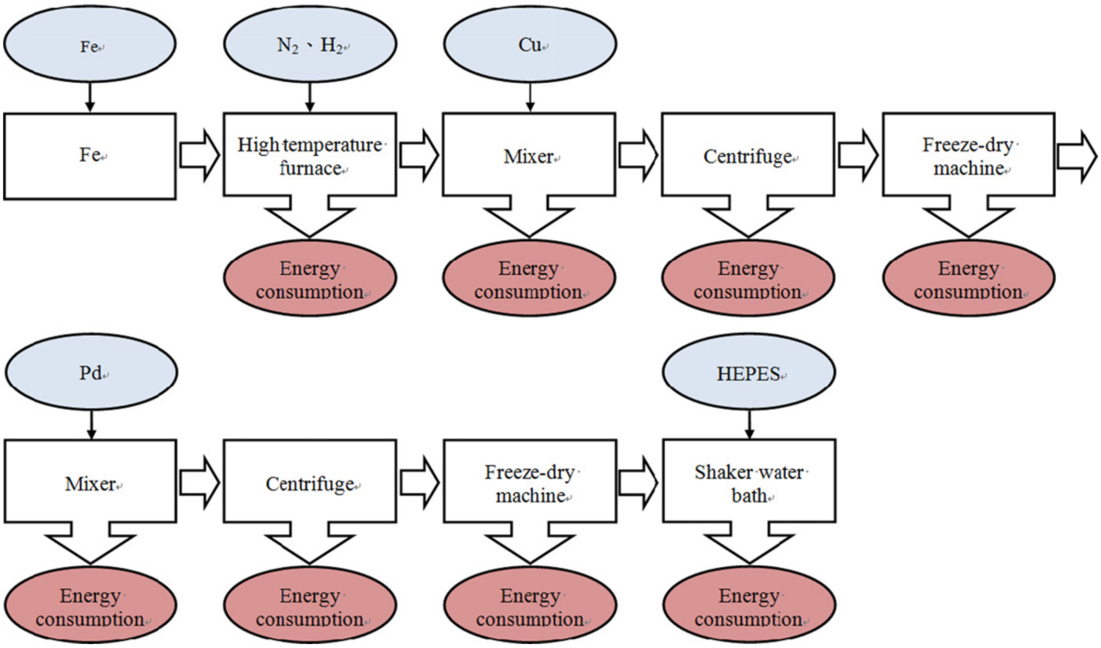

Fig. 5. Flow diagram for nitrate removal by Pd–Cu/FeO (Case E).

transportation, application, emissions, and waste treatment and disposal. The assessment was based on the removal of 0.325 kg nitrate, which was calculated using the maximum concentration of 121 mg/L of nitrate in the groundwater of Hualien County (Taiwan), with an assumed average well yield of 500 L/h. The treatment was spread out over a period of 24 h, allowing sufficient time for all 5 methods to remove all nitrates from the water. Since there is no full-scale reference plant using the Pd-based catalytic technologies, all 5 methods were investigated according to laboratory data. Despite the fact that some materials used in these 5 methods could be recycled or reused, the additional processing would add considerably to the associated time and cost. Additionally, reusing recycled material would lower the efficiency of the process; therefore, the use of recycled or reused material was not considered in this research.

3.1.2. *Inventory analysis*

As shown in Table 1, nineteen inputs associated with raw materials, energy consumption, waste discharge, and total costs were inventoried and analyzed. The inventory of total costs was carried out using the prices quoted by manufacturers for material costs, and the data published by Taiwan Power Company related to the quantity of energy consumed. The inventory of the laboratory equipments and instruments such as microwave reactors, refrigerated dryers, and centrifuges was conducted on-site, in laboratories. Most laboratory instruments handle 4–8 samples per batch operation; therefore, the total energy consumption is evenly allocated for each sample. With regard to total cost, Case A was the most expensive technology, nearly one order of magnitude higher than the other technologies. The main reason was that Case A consumed the greatest quantity of palladium and copper, and was the most energy intensive. Case C was the cheapest technology, but there was only a slight gap between Cases B and C. It can be examined from the flow diagrams for preparing the catalysts and the inputs in Table 1. The difference between Cases B and C was the catalyst preparation process in which Case B applied hydrogen-aeration and Case C added 0.04 mL/L of formic acid as the electron captor. For the

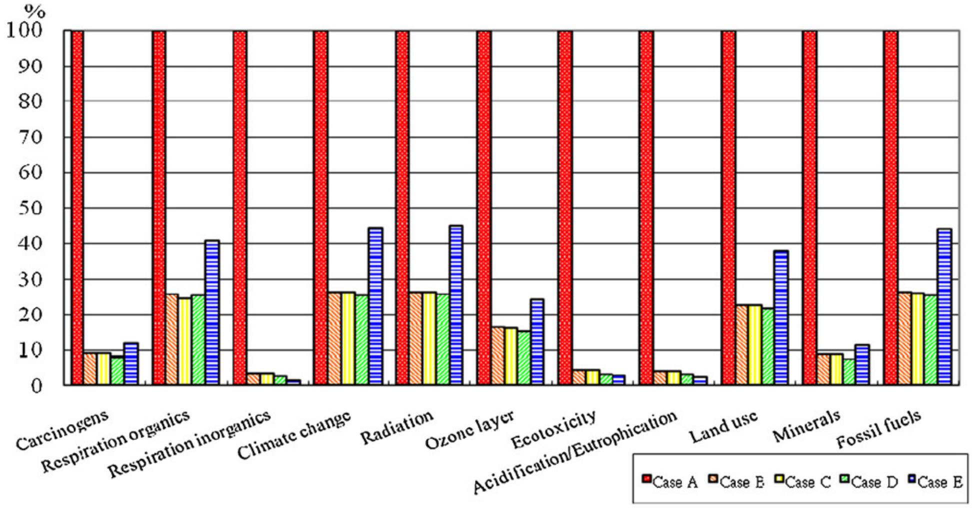

Fig. 6. Comparison using the Ecoeco-indicator 99 model of potential environmental impact for the five cases.

Table 2
Characterization of the different environmental impacts for five cases.

Impact category	Unit	Case A (Cu–Pd/TNTs)	Case B (H_2 + Pd–Cu/TiO_2)	Case C (Pd–Cu/TiO_2)	Case D (Pd/ZnO)	Case E (Pd–Cu/FeO)
Human health						
Carcinogens	DALY[a]	5.19E−03	4.74E−04	4.71E−04	4.32E−04	6.29E−04
Respiration organics	DALY	4.30E−05	1.11E−05	1.07E−05	1.11E−05	1.76E−05
Respiration inorganics	DALY	1.96E+00	7.34E−02	7.34E−02	5.40E−02	3.36E−02
Climate change	DALY	2.66E−02	7.06E−03	7.05E−03	6.86E−03	1.19E−02
Radiation	DALY	1.10E−03	2.91E−04	2.91E−04	2.85E−04	4.94E−04
Ozone layer	DALY	6.78E−06	1.12E−06	1.12E−06	1.06E−06	1.66E−06
Ecosystem quality						
Ecotoxicity	PAF[b] × m^2 × yr	1.04E+04	4.61E+02	4.60E+02	3.57E+02	3.12E+02
Acidification/Eutrophication	PDF[c] × m^2 × yr	3.79E+04	1.61E+03	1.61E+03	1.23E+03	1.00E+03
Land use	PDF × m2 × yr	1.87E+03	4.29E+02	4.28E+02	4.13E+02	7.08E+02
Resources						
Minerals	MJ surplus	1.82E+03	1.62E+02	1.60E+02	1.42E+02	2.08E+02
Fossil fuels	MJ surplus	1.23E+05	3.26E+04	3.24E+04	3.15E+04	5.46E+04

[a] Disability adjusted life years.
[b] Potentially affected fraction.
[c] Potentially disappeared fraction.

Cases D and E, their costs are in the middle range among all 5 cases and the difference between them is due to more laded Pd on Case D.

3.1.3. Impact assessment and interpretation

According to the potential environmental impact categories included in the Eco-indicator 99 model, the inventory were analyzed to express the relative contributions of substances to each category (Table 2). For all 5 cases, the impact of respiration inorganics and climate change were identified as having the most effect on the human health, while the impact of ecotoxicity, acidification/eutrophication, and land use on the ecosystem quality were about the same. Furthermore, the impact of fossil fuels was nearly two-order of magnitude higher than minerals in the resource depletion category. For each impact category, the impact of Case A was characterized to be the highest among the 5 cases. For comparing the relative impact among cases in each category, Fig. 6 presents the normalized results by setting the greatest impact of each category as a reference. The results indicate that Case A has a higher degree of impact on the various environmental indices than the other cases, with the differences exceeding the next highest case by more than double. Moreover, Case E was illustrated to have the next highest impact on respiration organics, climate change, radiation, ozone layer, land use, minerals, and fossil fuels categories. It can be attributed to higher electricity use and hydrogen-aeration from catalyst preparing process. But it had the best environmental performance in the respiration inorganics, ecotoxicity and acidification/eutrophication categories since it used the least quantity of Pd.

Table 3 indicates the results of each damage category based on the default weightings in the Eco-indicator 99 model. In the human health category, it is shown the primary contribution was the use of Pd for all cases except Case E. In ecosystem quality, the Pd use in Cases A–C was identified as having the most impact contributing 56.63–93.99%; and the consumption of electricity was the primary impact in Cases D and E. As for the damage of resource depletion, the use of electricity contributed the majority in all cases. In summary of the comparison of contributions of inputs to each damage category, the consumption of precious metals and energy were consistently shown in all categories for all cases that they are the highest impact. Palladium is scarce in nature; the earth's crust contains less than 1–10 μg/kg palladium; sea water contains less than 1 ng/kg palladium. Additionally, even a minute quantity of the metal causes eye and skin allergies. For these reasons, palladium is considered a significant environmental hazard. On the other hand, the electricity structure in Taiwan includes large fractions of

Table 3
Main impact factors in damage categories for five cases.

Damage category		Case A (Cu–Pd/TNTs)	Case B (H_2 + Pd–Cu/TiO_2)	Case C (Pd–Cu/TiO_2)	Case D (Pd/ZnO)	Case E (Pd–Cu/FeO)
Human health (DALY)	Palladium	1.928	0.058	0.058	0.039	0.006
		96.83%	71.17%	71.21%	62.58%	12.40%
	Electricity	0.062	0.023	0.023	0.023	0.04
		3.11%	28.56%	28.58%	36.89%	86.75%
	Others	0.001	0.0002	0.0002	0.0003	0.0004
		0.06%	0.27%	0.21%	0.53%	0.85%
Ecosystem quality (PAF × m2 × yr)	Palladium	47143.5	1414.3	1414.3	942.87	141.46
		93.99%	56.63%	56.68%	47.04%	6.99%
	Electricity	2857.07	1070.18	1070.18	1047.84	1865.63
		5.70%	42.85%	42.89%	52.28%	92.23%
	Others	158.47	12.81	10.74	13.65	15.68
		0.31%	0.52%	0.43%	0.68%	0.78%
Resources (MJ surplus)	Palladium	41176	1235.28	1235.28	823.52	123.55
		32.94%	3.77%	3.79%	2.60%	0.23%
	Electricity	82929.3	31063.1	31063.1	30414.7	54151.8
		66.34%	94.72%	95.29%	96.00%	98.82%
	Others	903.03	497.21	301.52	442.34	523.08
		0.72%	1.51%	0.92%	1.40%	0.95%

Table 4
Scores for the environmentally friendly evaluation.

Case	Case A (Cu–Pd/TNTs)	Case B (H_2 + Pd–Cu/TiO_2)	Case C (Pd–Cu/TiO_2)	Case D (Pd/ZnO)	Case E (Pd–Cu/FeO)
Environmental impact score (AHP weighting)	0.632	0.084	0.084	0.079	0.124
Cost score	0.603	0.067	0.059	0.137	0.133
Original impact scores	0.617	0.076	0.071	0.108	0.128
Normalization (based on Case C)	8.660	1.060	1.000	1.512	1.801

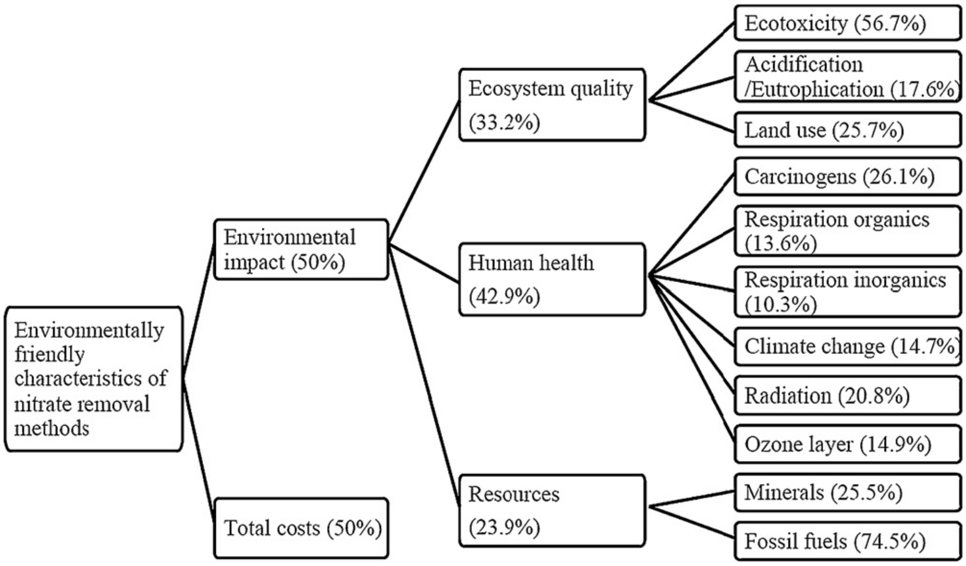

Fig. 7. Decision hierarchy structure and weighted factors for nitrate removal.

power plants using coal combustion resulting in high impact on climate change, acidification/eutrophication and fossil fuels categories. In Case A, the nano-scale TNT had a specific surface area 5 times greater (i.e. about 256 m^2/g) than TiO_2, and the quantity of palladium deposited on TNT in Case A (20%) was much higher than the quantities deposited on TiO_2 (Cases B and C) or on zero-valent metals (Cases D and E). The power consumption was also the highest among all cases. The technology used in Case A is considered to have the greatest environmental impact based on the Eco-indicator 99 assessment. Nevertheless, recycling and reusing precious metals was not considered for this study, the results related to environmental impact were based only on one-time use of the material.

3.2. *Analytic hierarchy process*

3.2.1. *Hierarchical structure*

The ultimate goal of the first hierarchy was set as “environmentally friendly characteristics”; the next level was divided into two parts with 1:1 weights for investigating the “environmental impact” and “total costs” in this research. The concept of equal weight refers to the approach of the eco-efficiency analysis by Shonnard et al. [18]. The criteria weighting under “environmental impact” was open to the collective decisions agreed upon by experts. The entire hierarchical structure is shown in Fig. 7.

3.2.2. *Consensus weight*

The consensus weights used in this research are based on a questionnaire investigation. We sent 20 questionnaires to experts who are members of the Chinese Institute of Environmental Engineering and have conducted relevant researches. Fifteen questionnaires were returned and analyzed, and 11 of them passed the consistency examination. The result of AHP questionnaires is shown in Table 5. As shown in Fig. 7, the decisions made by experts were 42.9% for human health, 33.2% for ecosystem quality and 23.9% for resources. The results indicate that the experts were most concerned with damage to human health, giving it the highest weight. If the results were expressed using absolute weights, the total economic costs and environmental impact would constitute 50% of each; the weights of indices under environmental impact were ranked according to their magnitude as: 9.4% for ecotoxicity, 8.9% for fossil fuels, 5.6% for carcinogens, 4.5% for radiation, 4.3% for land use, 3.2% for ozone layer that is the same as 3.2% for climate change, 3.0% for minerals, 2.9% for respiration organics that is the same as 2.9% for acidification/eutrophication, and 2.2% for respiration inorganics.

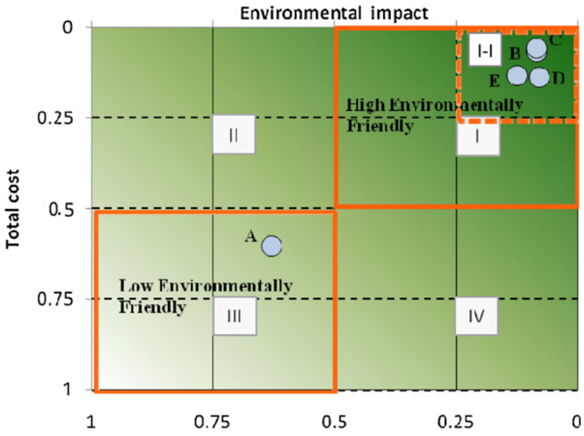

Fig. 8. Results of evaluation of environmental impact for five cases.

Table 5
Results of AHP questionnaires.

Experts impact catergories	1	2	3	4	5	6	7	8	9	10	11
Human health	0.443	0.714	0.333	0.540	0.540	0.229	0.625	0.443	0.122	0.143	0.528
Carcinogenic effects	0.335	0.299	0.166	0.258	0.358	0.083	0.324	0.100	0.340	0.124	0.210
Respiratory (organic)	0.121	0.054	0.142	0.124	0.128	0.074	0.212	0.065	0.162	0.076	0.115
Respirator (inorganic)	0.121	0.050	0.089	0.080	0.088	0.056	0.212	0.024	0.168	0.061	0.115
Climate change	0.044	0.104	0.296	0.087	0.033	0.455	0.051	0.322	0.207	0.227	0.054
Radiation	0.335	0.238	0.055	0.388	0.332	0.070	0.152	0.151	0.048	0.164	0.449
Ozone layer depletion	0.044	0.256	0.252	0.063	0.060	0.261	0.051	0.338	0.074	0.349	0.058
Ecosystem quality	0.387	0.143	0.097	0.297	0.297	0.696	0.238	0.169	0.558	0.429	0.333
Ecotoxicity	0.429	0.674	0.210	0.500	0.540	0.669	0.691	0.594	0.558	0.540	0.637
Acidification eutrophication	0.143	0.226	0.240	0.250	0.163	0.088	0.218	0.249	0.089	0.163	0.105
Land use	0.429	0.101	0.550	0.250	0.297	0.243	0.091	0.157	0.323	0.297	0.258
Resources	0.169	0.143	0.570	0.163	0.163	0.075	0.136	0.387	0.320	0.429	0.140
minerals	0.800	0.250	0.167	0.200	0.333	0.200	0.143	0.250	0.143	0.333	0.167
fossil fuels	0.200	0.750	0.833	0.800	0.667	0.800	0.857	0.750	0.857	0.667	0.833

3.3. The environmentally friendly assessment system

The results of LCA (Table 2) were normalized and combined with the consensus weights for the criteria in the AHP method, the impact scores expressing environmentally friendly features of the evaluated cases were compiled (Table 4) and are illustrated in Fig. 8. The results show that Case C, with the lowest impact score, is the most environmentally friendly technology, based on multidimensional evaluation based on the removal of an equal amount of nitrate. Conversely, Case A had the highest score, with and environmental impact score 8.5 times greater than that of Case C.

The degree of environmental impact determined in this research was obtained by offering equal weights to environmental impact and total economic cost, and combining these to obtain the overall results. Therefore, the cases located in the first quadrant in Fig. 8 were defined as “higher” environmentally friendly due to their lower cost and less adverse environmental impact. In other words, the cases located in the third quadrant in Fig. 8 then defined as less environmental friendly. We further divided the first quadrant into four sub-quadrants, in which Cases C, B, D, and E located (I–I) were defined as “highly” environmentally friendly. While Case A located in the quadrant III and defined as environmentally unfriendly. The 5 cases are ranked according to the degree of environmentally friendliness as C > B > D > E > A.

4. Conclusion

We carried out LCA for five catalytic denitrification methods using SimaPro 7.1 software and the Eco-indicator 99 evaluation method. The results reveal that the demand for the precious metal palladium is a major source of environmental burden, with the consumption of electricity as the next most important factor for evaluating environmental impact. Hence, seeking an alternative to palladium, and developing synthetic technologies that consumes less energy will be recommended for future study. In this research, the validity of data and information was examined using laboratory-scale nitrate removal technologies; the proportion of nitrogen removed and the optimal operating conditions were obtained using synthetic samples prepared by dissolving nitrates in pure water. Future field applications of the laboratory results must consider the actual quality of water, maturity of technology, and expected results. In addition, the environmental impact of batch nitrate removal technology may be over-estimated because the benefit of recovering and reusing the spent precious metal has not been included in the assessment.

Furthermore, environmentally friendly features and total economic costs were given the same weights based on the combination of AHP assessment results and the opinion of a panel of experts. The results reveal the following weights: 42.9% for human health, 33.2% for ecosystem quality, and 23.9% for resource, indicating that experts are more concerned about potential health damages of such treatment processes. Based on an equal amount of nitrate removal, and the evaluations of environmental impact and economic cost, the most environmentally friendly technology is Case C using palladium and copper covered titanium dioxide. The relative degree of environmentally friendliness for all 5 processes is Case C ($Pd–Cu/TiO_2$) > Case B (H_2 + $Pd–Cu/TiO_2$) > Case D (Pd/ZnO) > Case E (Pd–Cu/FeO) > Case A (Cu–Pd/TNTs).

Acknowledgements

The authors would like to thank the editor Gianluca Li Puma and two anonymous reviewers for their valuable comments and suggestions, which are very helpful in improving this paper.

References

[1] L.J. Puckett, Identifying the major sources of nutrient water pollution, Environ. Sci. Technol. 29 (9) (1995) 408A–414A.
[2] L.W. Canter, Nitrate in Groundwater, CRC Press, Boca Raton, FL, 1997.
[3] Y.H. Liou, S.L. Lo, C.J. Lin, W.H. Kuan, S.C. Weng, Chemical reduction of an unbuffered nitrate solution using catalyzed and uncatalyzed nanoscale iron particles, J. Hazard. Mater. 127 (2005) 102–110.
[4] A. Kapoor, T. Viraraghavan, Nitrate removal from drinking water – review, J. Environ. Eng. ASCE 123 (4) (1997) 371–380.
[5] M. D'Arino, F. Pinna, G. Strukul, Nitrate and nitrite hydrogenation with Pd and Pt/SnO_2 catalysts: the effect of the support porosity and the role of carbon dioxide in the control of selectivity, Appl. Catal. B: Environ. 53 (3) (2004) 161–168.
[6] Y.H. Hwang, D.G. Kim, H.S. Shin, Mechanism study of nitrate reduction by nano zero valent iron, J. Hazard. Mater. 185 (2–3) (2011) 1513–1521.
[7] M.I.M. Soares, Biological denitrification of groundwater, water, air, Soil Pollut. 123 (1) (2000) 183–193.
[8] M. Kumar, S. Chakraborty, Chemical denitrification of water by zero-valent magnesium powder, J. Hazard. Mater. 135 (1–3) (2006) 112–121.
[9] J. Li, Y. Li, Q. Meng, Removal of nitrate by zero-valent iron and pillared bentonite, J. Hazard. Mater. 174 (1–3) (2010) 188–193.
[10] G.C.C. Yang, H.L. Lee, Chemical reduction of nitrate by nanosized iron: kinetics and pathways, Water Res. 39 (5) (2005) 884–894.
[11] A. Devadas, S. Vasudevan, F. Epron, Nitrate reduction in water: influence of the addition of a second metal on the performances of the Pd/CeO_2 catalyst, J. Hazard. Mater. 185 (2–3) (2011) 1412–1417.
[12] S. Hörold, K.D. Vorlop, T. Tacke, M. Sell, Development of catalysts for a selective nitrate and nitrite removal from drinking water, Catal. Today 17 (1–2) (1993) 21–30.
[13] W. Gao, R. Jin, J. Chen, X. Guan, H. Zeng, F. Zhang, N. Guan, Titania-supported bimetallic catalysts for photocatalytic reduction of nitrate, Catal. Today 90 (3–4) (2004) 331–336.
[14] Y.H. Liou, G.J. Lin, S.H. Weng, H.H. Ou, S.L. Lo, Selective decomposition of aqueous nitrate into nitrogen using iron deposited bimetals, Environ. Sci. Technol. 43 (7) (2009) 2482–2488.
[15] A. Helland, H. Kastenholz, Development of nanotechnology in light of sustainability, J. Cleaner Prod. 16 (8–9) (2008) 885–888.
[16] M. Farre, J. Garcia-Montano, N. Ruiz, I. Munoz, X. Domenech, J. Peral, Life cycle assessment of the removal of diuron and linuron herbicides from water using three environmentally friendly technologies, Environ. Technol. 28 (7) (2007) 819–830.

[17] J. García-Montaño, N. Ruiz, I. Muñoz, X. Domènech, J.A. García-Hortal, F. Torrades, J. Peral, Environmental assessment of different photo-Fenton approaches for commercial reactive dye removal, J. Hazard. Mater. 138 (2) (2006) 218–225.

[18] D.R. Shonnard, A. Kicherer, P. Saling, Industrial applications using BASF eco-efficiency analysis: perspectives on green engineering principles, Environ. Sci. Technol. 37 (23) (2003) 5340–5348.

[19] R. Pineda-Henson, A.B. Culaba, G.A. Mendoza, Evaluating environmental performance of pulp and paper manufacturing using the analytic hierarchy process and life cycle assessment, J. Ind. Ecol. 6 (1) (2002) 15–28.

[20] P.T. Chiueh, S.L. Lo, C.L. Chang, A GIS-based system for allocating municipal solid waste incinerator compensatory fund, Waste Manage. 28 (12) (2008) 2690–2701.

[21] H.Y. Chen, Study on heterogeneous catalytic aqueous nitrate over Cu–Pd/titanate nanotubes, Master Thesis, National Taiwan University, Taipei, Taiwan, ROC, 2009.

[22] S.Y. Chen, Selective catalytic hydrogenation of nitrate to nitrogen on Pd–Cu/TiO_2 catalysts, Master Thesis, National Taiwan University, Taipei, Taiwan, ROC, 2009.

[23] Y.F. Tseng, Selective photocatalytic reduction of nitrate to nitrogen on Pd–Cu/TiO_2 catalysts, Master Thesis, National Taiwan University, Taipei, Taiwan ROC, 2007.

[24] I.C. Hung, Reduction of nitrate by zero-valent zinc and Pd/Zn Bimetallic Particles, Master Thesis, National Taiwan University, Taipei, Taiwan, ROC, 2009.

[25] W.Y. Tseng, Reduction of nitrate by catalytic bimetallic particles, Master Thesis, National Taiwan University, Taipei, Taiwan, ROC, 2006.

[26] J. Fava, R. Dennison, B. Jones, M.A. Curran, B. Vigon, S. Selke, J. Barnum, A Technical Framework for Life Cycle Assessment, SETAC, Washington, 1991.

[27] ISO 14040, Environmental Management, Life Cycle Assessment, Principles and Framework, 1997.

[28] M. Goedkoop, R. Spriensma, The Netherlands PRé Consultants, The Eco-indicator 99: A Damage Oriented Method for Life Cycle Impact Assessment, Methodology Report, 1999.

[29] A. Witoon, P. Pornpote, D.J. Watts, W. Koetsichai, LCA of spent fluorescent lamps in Thailand at various rates of recycling, J. Cleaner Prod. 16 (10) (2008) 1046–1061.

[30] C.J. Rydh, M. Sun, Life cycle inventory data for materials grouped according to environmental and material properties, J. Cleaner Prod. 13 (13–14) (2005) 1258–1268.

[31] T.L. Saaty, Decision making with the analytic hierarchy process, Int. J. Serv. Sci. 1 (1) (2008) 83–98.

[32] O.S. Vaidya, S. Kumar, Analytic hierarchy process: an overview of applications, Eur. J. Oper. Res. 169 (1) (2006) 1–29.

[33] C.C. Huang, H.W. Ma, A multidimensional environmental evaluation of packaging materials, Sci. Total Environ. 324 (1–3) (2004) 161–172.

[34] A. Kengpol, C.O. Brien, The development of a decision support tool for the selection of advanced technology to achieve rapid product development, Int. J. Prod. Econ. 69 (2) (2001) 177–191.

[35] T.L. Satty, The Analytic Hierarchy Process: Planning, Priority Setting and Resource Allocation, McGraw-Hill, New York, USA, 1980.