CEReS – Co-processing of Coal Mine & Electronic Wastes: Novel Resources for a Sustainable Future


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Abstract: Many coal mines produce waste which causes acid mine drainage (AMD) potentially resulting in severe environmental damage. This drainage can be treated, but most wastes will continue to produce such drainage for hundreds of years. At the same time, the pace of technological development means most electrical and electronic equipment becomes obsolete within a matter of years, resulting in the generation of vast quantities of electronic waste (e-waste). Where this cannot be recycled, it must be discarded. The CEReS project targeted the development of a co-processing approach to treat these waste streams to produce metals and other valuable products, while eliminating their environmental impact. This brings together two waste streams from opposite ends of the supply chain; turning each into a novel resource in a single, coherent ‘grave-to-cradle’ process. This industrial ecology approach is key to supporting a circular economy while securing the sustainable supply of critical raw materials. The project successfully elaborated a novel co-processing flow-sheet comprising: (i) the accelerated weathering of AMD-generating coal production wastes to generate a biolixiviant; (ii) the pyrolysis and catalytic cracking of low-grade PCBs to produce hydrocarbon fuel, a halogen brine a Cu-rich char; (iii) the leaching of base metals from the char using the biolixiviant; (iv) the reuse of the stabilised coal wastes; and (v) the recovery of valuable metal while concentrating precious and critical metals into enriched substrates. These individual process units were demonstrated individually at lab-pilot scale. The data were then used to validate the entire flow-sheet in an integrated process simulator. Finally an LCA approach was used to demonstrate the environmental benefits of the CEReS process over the status quo.

Keywords: AMD, Electronic Waste, Mine Waste, Pyrite, Co-processing, Industrial Ecology
1. Introduction

Poland is Europe’s largest producer of hard coal, and coal is used in the generation of approximately 80% of the country’s electricity. As a result of this mining activity ~25 Mt/y coal production waste is generated, much of which contains pyrite. While 90% is reused in civil and geotechnical engineering, there are problems where this material is subject to weathering. Tauron Wydobycie own the Janina & Sobieski coal mines and hold the largest coal reserves in Poland. These mines produce sulfidic wastes, much of which are currently stock-piled. A long-term solution is needed.

EU reliance on importation of critical raw materials (CRM) is well-documented. At the same time, the importation of electronic and electrical equipment (EEE) is an “open-loop”, representing an important reserve of CRM. Comet Traitements (Belgium) cover the entire recycling and valorisation chain. Currently they recover 97.8 % of end of life vehicles for reuse. They developed a pyrolytic process to treat the organic fraction of light shredder residues (LSR) producing hydrocarbon fuel, carbon-rich concentrates and metal-bearing char.

The CEReS process brings together these two waste streams – AMD-generating coal production wastes and waste EEE, in an example of industrial ecology. The technical feasibility of the individual unit processes will be assessed at laboratory/pilot scale before integration in silico. An LCA approach will be used to evaluate the environmental benefits of the CEReS process compared to the business as usual, do-nothing scenario.

The proposed co-processing flow-sheet is summarised in Figure 1, and can be considered in four parts: (i) AMD-generating coal mine wastes are recovered from existing mine waste dumps or during ongoing production. PCBs (shredded) are bled off from existing WEEE or e-waste handling streams; (ii) A catalytic cracking circuit is used for pre-treatment of the PCBs resulting in a metal-rich char. Catalytic cracking partially converts the organic fraction into a liquid fuel. A quenching process removes halogens (mostly bromine) as a saleable brine. The low temperatures and reducing conditions prevent the formation of dioxins while maximising product recovery; (iii) A bioreactor system is used to oxidise sulfide minerals in the coal wastes (effectively accelerating AMD production), resulting in the production of a ferric iron-sulfuric acid lixiviant. This is used to leach base metals from the PCB char; and (iv) Valuable metals are recovered from the pregnant leach solution (PLS) by appropriate downstream process. Precious metals, lead and tin will report to, or remain in, the solid leach residue.

![Figure 1: Simplified overview of the proposed CEReS co-processing flow-sheet.](image-url)
2. Results

2-1. Raw Materials

**Coal production waste.** Jig and spiral tails from Janina (JJT and JST, respectively) and Sobieski (SJT and SST, respectively) were extensively characterised. Detailed mineralogical analysis necessitated the development of a novel QEM-SCAN protocol. The Janina spiral tails were selected for the project due to their high pyrite content (approx. 12%). However, subsequent QEM-SCAN analysis indicated limited liberation of the pyrite (25-30%). Chemical and biokinetic tests confirmed that the selected material was acid-generating. However, far less acid was produced given the amount of sulfide oxidised (in both AMD tests and bioleaching experiments); this “missing acid” has yet to be explained.

**Scrap printed circuit boards.** The Polish WEEE market was analysed, and three categories of scrap PCBs were identified: high-, medium- and low-grade. The low-grade material was selected, but analysis found it was in fact a mixture of PCBs (~35%) and other parts of WEEE. Therefore, for the development of the project, Comet used low-grade PCBs from their own stocks. This material was analogous to the scrap PCBs available on the Polish market. The CEReS project focussed primarily on the recovery of copper, but further work should be undertaken to recover precious metals.

Finally, a database was created cross-mapping the occurrence of suitable mine wastes (dumps and mining operations) with WEEE handling facilities. This allows the identification of geographically suitable potential locations for a CEReS process.

2-2. Pyrolytic Cracking

The pyrolytic process was successfully adapted to scrap PCBs. Approximately 18% of the PCB mass could be converted to combustible hydrocarbons, the calorific value of which is sufficient to power the pyrolysis reactor at industrial scale. Approximately 75% of bromine content was released from the solid residue of the pyrolysis into the quench water, to produce a saleable Br product. The metal-bearing char from the process was split into three size fractions: coarse (>8 mm, 6.2% Cu), mid (0.075-8 mm; 41.9 % Cu) and fine (<0.075 mm; 4.2% Cu). The mid fraction was sufficiently Cu, Au and Ag-rich that it can be sold directly to existing metal refiners. The fine fraction can be disposed of conventionally while the other fractions are the target for leaching using the biolixiviant.

2-3. Lixiviant Production and Char Leaching

**Bioleaching and lixiviant production.** Two enrichment cultures at 30°C (TW30) and 48°C (TW48) were compared to two consortia of proven bioleaching ability: meso (30°C) and BRGM-KCC (42°C). Following extensive tests, the TW48 culture at 48°C was selected. A suitable biolixiviant could be produced from the coal production waste, (~10 g/L Fe³⁺).

**Char leaching.** A stirred-tank reactor (STR) and a rotating drum reactor (RDR) were tested with various operating and char pre-treatment options. The best performance was obtained with an STR set-up, but this was not suitable for the larger (2-8 mm) char fraction. Conversely, the RDR was not suitable for leaching the finer fractions as the char was not retained within the inner mesh drum. Therefore, the RDR was selected to leach the coarse fraction and an STR for the fine fraction.
2-4. Metals recovery and process residues

**Metal recovery.** The highly selective Acorga reagent was chosen for solvent extraction (SX) with a two-stage mixer-settler configuration followed by electrowinning (EW). Within the scope of the project, it was only possible to valorise the copper, with the precious metals sold to existing refiners within the leached char residue.

**Process residues.** Environmental toxicity tests indicated that the leached char was less of a threat to the environment than the PCBs prior to treatment. The sulfide component of the coal production waste was almost completely removed following bioleaching. Accordingly, the maximum potential acidity (MPA) was reduced. The amount of sulfate produced during net acid generation tests (NAG) suggested the material was non-acid-forming, but the pH of the solution became acidic and negative acid neutralising capacities (ANC) were measured. This suggested that the material leaches acid, but not through the oxidation of residual sulfides. It may be that the “missing” acid is adsorbed to the material and may desorb in the environment.

Four options were considered for the reuse of the leached coal waste. The addition of leached material (15% initial mass) during brick production produced satisfactory bricks with slightly elevated compressive strength. Incorporation into cement products (15%) had no detrimental effects and slightly improved compressive strength. Concrete polymer products could be made from the waste alone. However, this required a long bonding time rendering this option for reuse impractical. The leached waste was used in for the production of granulates. The granulates produced were of high strength and tests confirmed that leachates did not exceed permissible contaminant levels. Therefore, such granulates can be safely used in underground mining techniques and civil engineering.

2-5. Economic and Environmental Assessment

**Process integration through simulation.** A global simulator was created using USIM PAC, allowing an economic assessment and evaluation of alternative flowsheet options. Reprocessing coal mine waste using CEReS would cost approximately 93 €/t. However, the current simulation assumes that the stabilised wastes have no value, which may not be the case if, for example, the ceramic or concrete products could be sold. Furthermore, the majority of the PCB value is in the precious metals and the current flowsheet does not include a process for their valorisation. This will be further developed in the future.

**Life cycle analysis (LCA).** A comprehensive LCA of the proposed flowsheet revealed that CEReS was more environmentally friendly than the current “do nothing” scenario in almost all categories; the exception was categories linked to fossil fuel use. This is due to the fact the CEReS is a relatively energy-intense process and Poland’s (the case study country) energy mix is carbon-heavy. Nevertheless, the environmental benefits of CEReS are clear.

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