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Life Cycle Assessment and Environmental Profile Evaluations of High Volumetric Efficiency Capacitors

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Abstract

High volumetric efficiency capacitors are found in all smart electronic devices, providing important applications within circuits, including flexible filter options, power storage and sensing, decoupling and circuit smoothing functions. Multilayer ceramic capacitors (MLCCs) hold the major market share but tantalum electrolytic capacitors (TECs) provide a viable alternative if higher breakdown strengths are required. The reduced costs, smaller dimensions suitable for space-constrained electronic circuits, exceptional high-frequency characteristics, higher reliability, ripple control and longevity, however, are driving the market to replace TECs with MLCCs wherever possible. To date, no current research regarding the transition from TECS to MLCCs has been conducted from an entirely environmental viewpoint. This article identifies, quantifies, ranks and compares the environmental impacts of the MLCC and TEC supply chains using an integrated hybrid life cycle assessment framework. Three recovery methods: incineration; hydrometallurgy and pyrometallurgy are considered in the overall impact assessment. Electrical energy consumption during fabrication alongside the use of nickel paste are the major environmental hotspot for MLCCs. The high proportion of tantalum in TECs results in an overall greater environmental impact in comparison with MLCCs, due to intensive extraction, processing and purification requirements of tantalum. Of the three recovery methods, the hydrometallurgy process offers the least environmental impact for both MLCCs and TECs. Overall, the current work shows that while the industry led transition from TECs to MLCCs offers both an operational and functional edge, it is also an environmentally intelligent move. Intervention options that can further drive down the environmental impacts of MLCCs are also proposed such as a reduction in the reliance of MLCCs on rare earth elements and Cu external electrodes in some designs and material recovery.

Keywords

Capacitors, Multilayer Ceramic Capacitors, Tantalum Electrolytic Capacitors, Functional Materials, Hybrid Life Cycle Assessment, SCEnAT

Nomenclature

A/DC	Alternating/direct current	SCEnAT	Supply Chain Environmental Analysis Tool
AEC	Aluminium electrolytic capacitor	TEC	Tantalum electrolytic capacitor
AP	Acidification potential	WEEE	Waste electrical and electronic equipment
ASM	Artisanal and small scale mining	X7R	MLCC specification
BOM	Bill of materials	A	Technical coefficient of the IO matrix
EC	Electrolytic capacitor	E_i	Emissions intensity
(E)IO	(Environmental) Input Output	$kgCO_2$-eq	kg of CO_2 equivalent
EP	Eutrophication potential	kVA	Kilo-volt-amps
ESR	Equivalent series resistance	kWh	Kilowatt hour
DCB	Dichlorobenzene	I	Identity matrix
GWP	Global warming potential	MHz	Mega hertz
HTP	Human toxicity potential	MJ-eq	Mega joule equivalent
LCA	Life cycle assessment	nm	Nano meters
LCI	Life cycle inventory	Q	Quantity of a material or process
MLCC	Multilayer ceramic capacitor	tanδ	Dissipation/power factor measurement
(W)PCB	Waste printed circuit board	W	Watts
POCP	Photochemical ozone creation potential	wt%	Weight percent
REE	Rare earth element	μF	Micro farads

1. Introduction

The use of functional materials in product and device development underpins many aspects of modern life through energy generation and storage devices, information and communications technology, multicomponent sensors, healthcare, military defence and transportation. Modern society has witnessed tremendous growth and development through the discovery and applications of functional materials and semiconductor devices[1]. One specific area where the use of functional materials has made new applications possible is the fabrication of capacitors. A capacitor is a passive electrical component which possesses two terminals for energy storage within an electric field. Their capacitance is measured in farads (F) and is the ratio of the electric charge to the voltage difference between the two electrical conductors separated by a dielectric. There are numerous different types of capacitors including aluminium electrolytic capacitors (AECs), aluminium organic polymer capacitors, ceramic capacitors, single layer ceramic capacitors, multilayer ceramic capacitors (MLCCs), array capacitors, tantalum electrolytic capacitors (TECs) and supercapacitors. By identifying different attributes such as capacitance, rated voltage, operating temperature range and dimension, capacitors can be selected for different types of applications.

The importance of such devices cannot be underestimated. Modern society depends on a number of devices for which capacitors are used; the functional materials industry currently boasts of a world market size in excess of $4 trillion with a growth rate of 4.8% per annum[1]. The UK alone accommodates substantial cluster of manufacturers and end users of functional materials devices such as capacitors, production of capacitors in the UK reached over €1 million in 2013[2]. Given that the fabrication of products such as volumetric efficient capacitors rely heavily on raw materials which have geopolitical, geological and environmental constraints[3-5], the importance of tracking their environmental and social profile cannot be overemphasised.

Innovations in consumer electronics inevitably lead to the generation of waste electrical and electronic equipment (WEEE). Capacitors are soldered onto printed circuit boards (PCBs) and are a vital component of electronic circuits and therefore contribute to the 50 million tonnes of WEEE produced each year[6, 7]. With an annual growth rate of 3-5% per year, WEEE is thought to be one of the fastest growing waste streams in the world[8]. Currently, there is limited environmental profile assessment of capacitors in their various forms, Wang and Xu[6] submitted that a mature recycling technique is yet to be developed for capacitors and other electronic components although hydrometallurgy and pyrometallurgy can be used for precious metal recovery[9].

To this end, the current work presents a methodologically robust lifecycle assessment (LCA) of two representative capacitors, namely Tantalum Electrolytic

Capacitors (TECs) and Multilayer Ceramic Capacitors (MLCCs). This allows us to define and address environmental hotspots within the supply chain as well as sustainability issues that are essential for future development of these capacitors, given their wide array of applications. Research has not yet been published which highlights environmental impacts of both type of capacitor and therefore a comparison is yet to be made. Although there are numerous types of capacitors, the overall aim of the current work is to compare two main types that can be employed as immediate replacement and substitutes for similar applications. In this regard, MLCCs hold the major market share but tantalum electrolytic capacitors (TECs) provide a viable alternative if higher breakdown strengths are required, hence the trend to replace TECs with MLCCs where possible. Also, TEC turnover is 75% of the dollar compared to MLCCs placing them second in the capacitor industry in terms of units and value[10]. Moreover, there is a lack of availability of detailed life cycle inventory (LCI) data for all types of capacitors, as such, the consideration of all types of capacitor for LCA is beyond the scope of the current work. This work therefore provides a novel and important insight into the environmental impacts of the production of MLCCs and TECs at a laboratory scale. The results can be directly translated to the day to day production of each capacitor type and can serve as a viable tool in design decision making process.

1.1 The switch from electrolytic capacitors to multilayer ceramic capacitors

For applications requiring large capacitance (e.g. smoothing), both aluminium electrolytic capacitors (AECs) and TECs have been adopted. Difficulties in miniaturisation of these capacitors, coupled with significant self-heating problems from ripple currents has hampered their applications in a number of space-constrained electronic circuits. These challenges prompted the development of MLCCs. MLCCs were first adopted in a number of niche electronic applications as their capacitance was comparatively low, thus confining their use to filter and high-frequency circuits[11]. However, in recent years, with advances in technology for the multi-layering of dielectric materials, large-capacitance MLCCs have been fabricated, enabling the replacement of electrolytic capacitors (ECs) in a number of applications[12]. Their small dimensions, high capacitance, high reliability and exceptional high frequency characteristics find them now utilised in mobile phones (Figure 1), laptops and cars[12, 13].

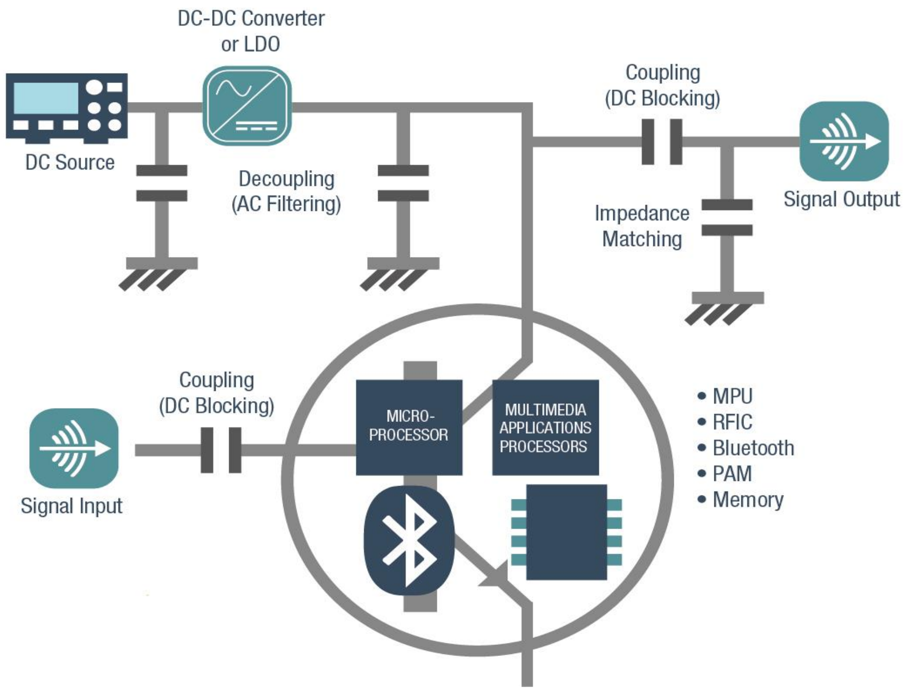
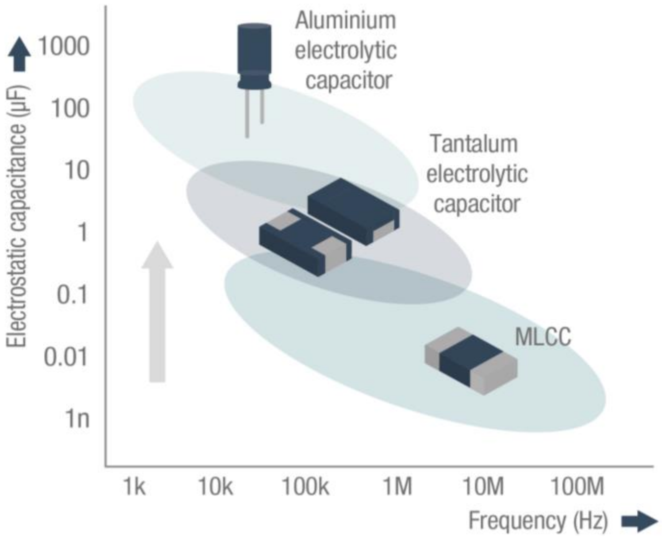
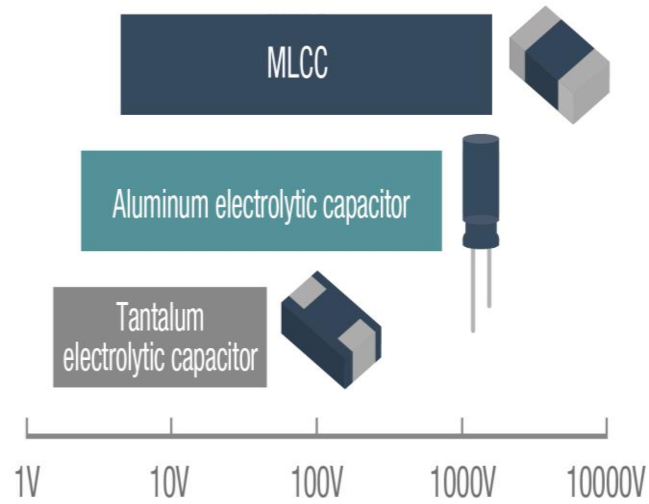


Figure 1: Schematic illustration of the application of MLCCs in mobile phones. As indicated, MLCCs can be used for coupling (e.g. DC blocking) and decoupling (AC filtering) as well as impedance matching.

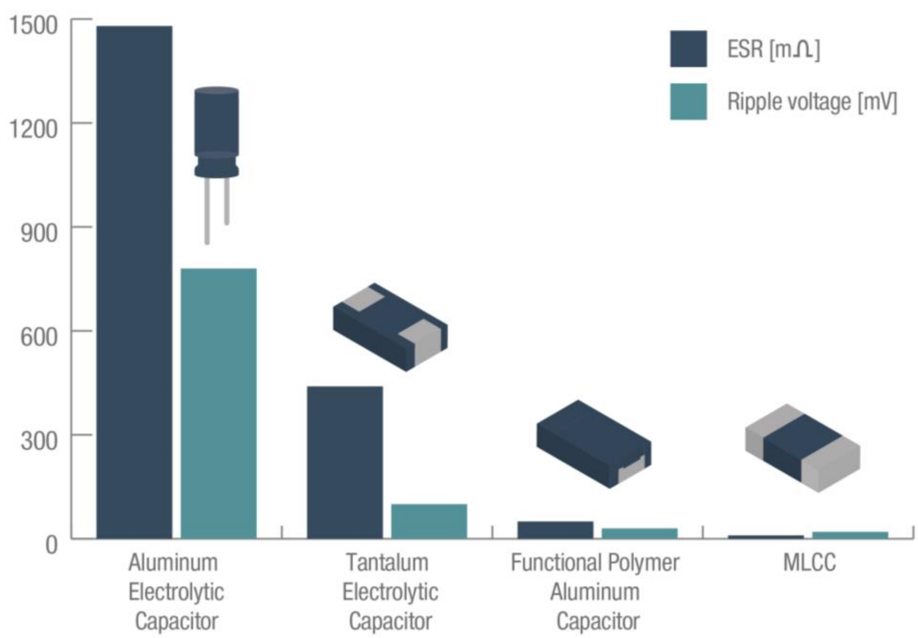
By 2020, ~3 trillion MLCCs per year will be required to fulfil the demand for computers, smart phones and computerised consumer electronics[14]. TECs[10] also have high reliability, high volume efficiency and good temperature characteristics and consequently compete in the same market as MLCCs[15]. Although the switch from TECs to MLCCs offers the aforementioned advantages, their shortcomings lie in the large rate of change in capacitance as a result of temperature and DC bias. MLCCs also possess low equivalent series resistance (ESR) which can cause adverse effects that may lead to anomalous oscillations in power supply circuits[12]. Figure 2 illustrates trends towards the switch from ECs to MLCCs.



(a) Frequency band and range of capacitance



(b) Rated voltage range

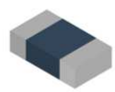
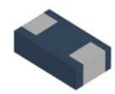
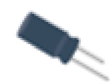


(c) Relationship between ESR and ripple voltage

Figure 2: Trends towards the switch from ECs to MLCCs. (a) replacement of ECs (AEC and TEC) due to the advent of large-capacitance MLCCs; (b) rated voltage range of sample capacitors. MLCCs possess higher voltage ratings in comparison with ECs. It is also endowed with longevity and superior reliability; (c) relationship between ESR and ripple voltage. A lower ESR allows the ripple voltage to be maintained to a smaller amount, an attribute of MLCCs which enhances its optimal performance as a replacement for ECs.

MLCCs and TECs have different manufacturing materials and processes, but the same function in power correction and smoothing a digital circuit[16]. Companies including Kemet[17], YAGEO[18] and academic articles (e.g. Huang et al.[19]) have discussed the implications of replacing TECs for MLCCs with the latter concluding that total replacement, at this time, would not be feasible due to capacitance and temperature limitations. Table 1 provides a summary of the key differences between aluminium and tantalum ECs and MLCCs.

Table 1: Key functional difference between MLCCs and ECs (TECs and AECs)

	MLCC 	**TEC** 	**AEC** 
Technical features	• Small size, low profile form factor • Very large capacitance • High reliability • Longer lifespan • Low equivalent series resistance (ESR) • No polarity • High voltage rating	• Large capacitance • Advanced DC bias characteristics	• Large capacitance • Less expensive
Caution in application phase	• Large change in capacitance due to temperature and DC bias • Low ESR constitute an advantage but may cause oscillation problems in power circuits when too low	• Comparatively high ESR, significant self-heating because of ripple currents • Low voltage rating	• Large form factor • Short lifespan in environments with high temperature • High ESR, significant self-heating because of ripple currents

As highlighted above, switching from MLCCs offers a number of benefits including small size (due to the miniature and low-profile form factor), improved reliability, ripple control and longevity[12]. However, caution must be taken due to the low ESR attributes of the MLCC which can have adverse effects leading to anomalous oscillations and anti-resonance[11]. The lifespan of a typical AEC is estimated to be about ten years because its capacitance decreases as the electrolytic solution dries up. MLCCs however do not suffer from these limitations because they contain almost no components and as such, they are endowed with longer lifespan. The increasing desire to adopt primary large-scale integration and integrated circuit components within electronic devices couple with the trend towards low voltage in power supplies which powers these components invigorated the race to replace ECs with MLCCs. Additionally, the consumption of power has increased considerably in line with the progression of multi-functionality in electronic devices and the trend towards the use of high current continues. This trend towards high current and low voltage in electronic devices has been further enhanced due to the replacement of ECs with MLCCs[12]. The enormous advantages of the switch from ECs to MLCCs can therefore not be overemphasised.

While it is clearly well-established that the transition from ECs to MLCCs offers both an operational and functional edge as highlighted in the aforementioned examples, there is currently no research regarding such transitions that has been conducted from an entirely environmental viewpoint. For years to come, the manufacturing of the capacitors under consideration would continue in order to fill important human needs. As such, an understanding of their environmental profile is therefore paramount. Such an understanding will provide manufacturers of capacitors and allied professionals with an optimal and reliable input into the design process that is informed by environmental considerations. In the subsection that follows, the need to conduct LCA of volumetric capacitors is provided.

1.2 Towards life cycle assessment of volumetric efficiency capacitors

As highlighted in the preceding paragraphs, a great deal of progress and improvements based on the performance characteristics and functional aspects of volumetric efficient capacitors have been recorded. Yet, despite the importance and volume of capacitors in today's electronics, there are no LCA studies immediately available to track the progress recorded from a purely environmental perspective. At the moment, only one LCA for a MLCC was found which contained information limited in scope; the results may be commercially sensitive and therefore remain unpublished[20]. The Ecoinvent database[21] holds a dataset for “capacitor production, tantalum-, for through-hole mounting”. There is also information regarding generic capacitors in the Ecoinvent database which is referenced as ‘capacitor, for surface mounting’. However, none of these sources of environmental information have presented a detailed cradle-to-grave analysis of the entire fabrication route for comparison between TEC and MLCC. In an era where environmentally-sensitive manufacturing procedures are monitored with greater focus and attention, this is an important gap to fill given the increased global awareness of environmentally benign design and the strong relationship between global warming and CO_2 emissions.

The role of LCA to evaluate whole-life environmental impact of capacitors is crucial, as this can play an important function in the early stages of their design process. This is particularly important given the vital functions that capacitors perform in many devices and the fact that their production will continue to grow, especially considering that product supply chains are networked with complex production systems[22, 23] and unpredictable and ever increasing consumption patterns[24, 25].

1.2.1 Summary of contributions and novelty

The novelty and contribution of this paper is summarised as follows:

a) The current work presents the first and comprehensive comparative LCA of two representative volumetric efficient capacitors namely MLCCs and TECs with the view to: (i) provide information to be used at the design phase of capacitors with regards to the environmental and health impacts of each component and (ii) highlight environmental hotspots and recommend mitigation strategies and intervention options for future designs. It is intended that the analysis presented provides manufacturers of capacitors and allied professionals with an efficient and reliable input into the design process that is informed by environmental considerations.

b) The application of hybrid LCA framework to identify supply chain hotspots in the environmental profile of High Volumetric Efficiency Capacitors. The work demonstrates the analytical capability of LCA for the environmental impact assessment of new device versus existing device across multiple environmental metrics. In particular, it highlights the fact that the replacement of ECs with MLCC is an environmentally intelligent move. This is an important information for designers and manufacturers of capacitors.

c) Demonstration of the important application of integrated hybrid LCA to a strategic manufacturing procedure which allows equipment and device designers, as well as policy makers, to make informed decisions regarding the environmental consequences of substitute materials, designs, manufacturing processes and application.

In light of the above, the remainder of the paper is organised as follows. In Section 2, a succinct literature review detailing materials composition/ requirements and the recyclability potentials of capacitors is provided. A brief description of the steps involved for the fabrication processes of laboratory-based MLCCs and TECs are presented in Section 3. Details of the general methodological notes and theoretical formulations underpinning the Supply Chain Environmental Analysis Tool (SCEnAT) based on integrated hybrid LCA model is provided in Section 4. In Section 5, the key findings of the results are analysed and discussed leading to the summary and concluding remarks in Section 6.

2. Literature review

2.1 The role of capacitors in improving energy efficiency of systems

Due to the depletion in fossil fuel reserves and the ensuing climate change impacts as well as the need to pursue complete energy independence, the importance of developing efficient systems to support climate change mitigation initiatives have become more apparent[26, 27]. This has led to increased awareness about conservation of energy, prompting the need to utilise available energy in an efficient manner through the use of energy efficient devices[28]. To achieve this, there is the need to convert conventional systems into energy efficient systems. Capacitors can play a vital role in achieving this goal as they constitute an integral part in constructing energy efficient systems[29, 30].

As with almost all electronic components, the automotive systems put capacitors into extensive use. In fact, the rising adoption of cars utilising alternative propulsion technologies where the management of electrical current and circuits is becoming more important has led to further expansion in the role of capacitors. Innovations into supercapacitors have equally rendered these devices suitable for use in electric vehicles and plug-in hybrids, supplementing and in some instances replacing batteries[31, 32]. Throughout the automotive subsystems of all types of cars, different types of capacitors can be found. For example, AECs are used in subsystems like window wipers, air conditioning as well as motors used for automatic windows, seats and other applications[33]. They are also used in important safety and control systems like power steering, breaking systems and airbag controls, engine control units for battery controls and lots more[33]. Furthermore, for smooth grid integration of large-capacity renewable energy sources (e.g. solar and wind energy) and use of large-capacity electrical energy storage, capacitors will play a vital role towards an energy efficient system. Figure 3 illustrates a variable-slip induction generator where a capacitor is used as a reactive power compensator.

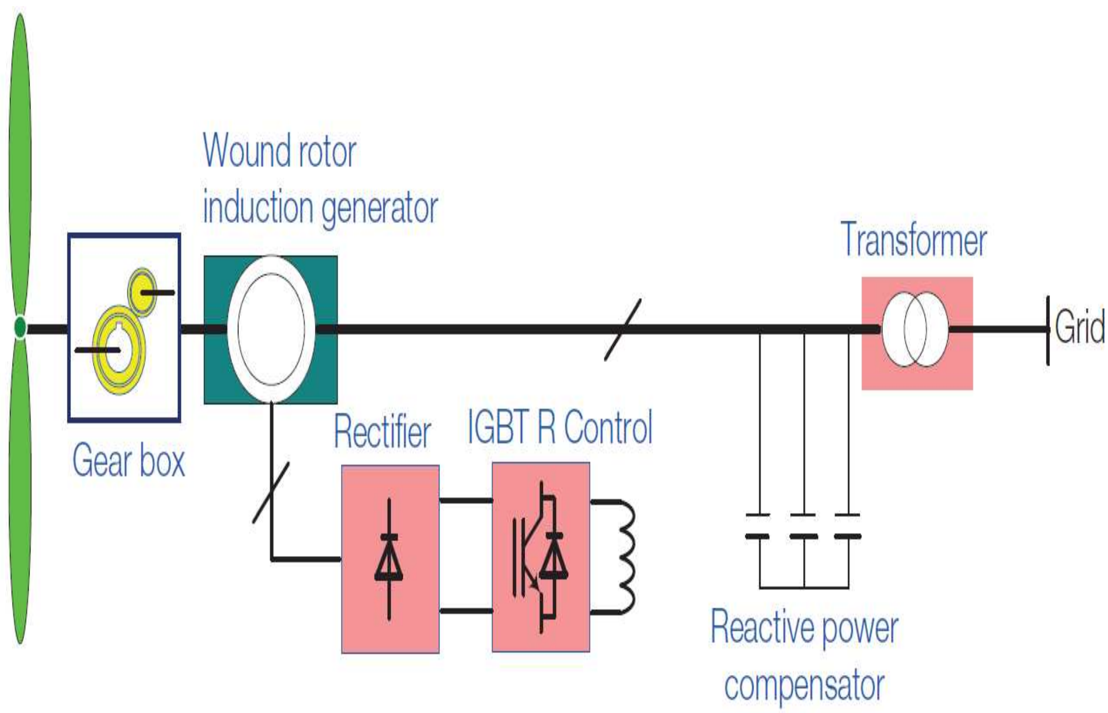


Figure 3: Topology of a variable-slip induction generator where a capacitor is used as a reactive power compensator grid integration of wind energy, adapted from International Electrotechnical Commission[34].

In order to conserve the use of energy, industries are rapidly changing to energy efficient equipment and drives such as DC drives, variable speed AC drives, uninterrupted power supply and energy efficient lamps. Although these devices can facilitate the use of energy in an efficient manner, they have the tendency of reducing the power factor of systems through the injection of harmonics which can lead to overall decrease in efficiency of the systems. For instance, the need for accurate and automatic control in systems and devices has led to the development of electronic controls. However, some of these devices requires switched mode power supply which draws current over a part of each half cycles thereby reducing the power factor. A number of numerous examples of reduction in power factor is available in electronic literature.

In an electrical system, low power factor constitutes a disadvantage given that it decreases the overall efficiency of the system whilst affecting the operability of other associated devices. Capacitors play a vital role in improving power factor[33, 35]. Improved or higher power factor leads to overall reduction in load current and power loss; improvement in efficiency of the system; reduction in KVA rating of the device whilst enhancing better utilisation of such device; better and improved voltage profile; less voltage fluctuations and increased stability[36, 37]. By using capacitors to achieve the aforementioned advantages, overall improvement in energy consumption pattern and efficiency of such systems can thus be guaranteed. More importantly, capacitor itself is an energy efficient device given its low power loss and overall efficiency of roughly 99.9%[38, 39].

2.2 Capacitor Types

Capacitors differ from other types of energy storage devices in that they are passive electrical devices that store small quantities of electrical energy, as opposed to electrochemical devices which produce energy such as batteries and fuel cells[16]. Storage of energy ranges from multiple terawatts hours held for years in chemical compounds, to watt hours held in capacitors for mere seconds[40]. The basic structure of a capacitor involves an insulating layer separating a minimum of two electrical conductors. Charging leads to the storage of electricity in the dielectric insulator which can be made of a ceramic, glass or polymer[16]. Supercapacitors, also known as electric double-layer capacitors and ultracapacitors, function in the gap between batteries and conventional capacitors as they have the characteristics of both energy storage types but are still only suitable for short term storage of energy[16, 41]. Furthermore, supercapacitors differ from capacitors in that they also have a porous membrane separator incorporated into the structure and utilise nano-scale materials to increase surface areas which increases capacitance[16].

Capacitor characteristics determine their use in different applications. For example, hybrid energy storage systems, combining electric double layer capacitors with lithium battery technology, are durable and have high power densities, focussing research on their optimum material design[42]. Electrolytic capacitors utilising aluminium, tantalum or niobium, have been developed over the last 120 years and are used in computer motherboards and larger power supplies with capacitance ranging from 1μF to 2.7F[10, 43]. Ceramic capacitors, such as multilayer ceramic capacitors (MLCCs), are capable of quickly charging and discharging with a high power density[44]. The specifications for X7R MLCCs require a minimum operating temperature of -55°C up to a maximum operating temperature of 125°C, with a percentage of variation in capacitance of ±15% across that temperature range. With these parameters in place, work focusses on producing a range of materials to suit these requirements[45].

2.3 Materials for capacitor fabrication

X7R MLCCs (with dielectric specified to standard 198 of the Electronic Industries Association) are one of the most common types of MLCC and use barium titanate (BT) as the dielectric component due to its high dielectric constant and low dielectric loss in the MHz frequency range[46, 47]. For this application very thin $BaTiO_3$ layers are required which are produced from uniform particles, typically 100-200nm in size. These powders are produced in several ways such as hydrothermal synthesis and solid-state reaction between barium carbonate ($BaCO_3$) and titanium dioxide (TiO_2)[48]. The addition of a rare-earth element, such as dysprosium (Dy) or holmium (Ho), to BT is known to maintain high insulation resistance for long periods, enhance temperature stability and therefore permit the use of thinner dielectric layers that have longer in-use life spans[49].

Dy is a rare earth element (REE) (one of 17 metallic elements with similar chemical characteristics; other REEs include yttrium, lanthanum and gadolinium), which is added to BT as its oxide, Dy_2O_3[50]. Critically, MLCCs utilize 2-3 wt% of either Dy, Ho, or Erbium (Er) oxide of which there is severe scarcity; the Department of Energy in the USA regards Dy as the number one most critically endangered element[51]. Contrary to what their name suggests, REEs are not rare in abundance but are difficult to obtain in economically feasible concentrations. Additionally, the separation and refining processes are challenging and hazardous for the environment[3, 52].

From 1965 to 1985, Mountain Pass, California produced the majority of the world's rare earth elements with Australia a major producer into the 1990s. More recently, China has been able to produce rare-earth elements more economically than in other regions causing the closure of financially unviable mines in America and Australia[13]. Two mines (one American-owned and one Australian-owned) became fully operational in 2013, potentially challenging the Chinese monopoly on the rare-earth market and addressing supply chain issues[53]. To illustrate the reliance on Chinese production, the 2010 European Commission report on 'Critical raw materials for the EU' stated that 90% of rare earth metals were produced in China but by 2014 the 'Report on Critical raw materials for the EU' Heavy rare earth elements (of which Dy is classified) recognized that this had increased to 99%[3-5]. Despite the domination in production (and restriction of exports) by China, Dy annual demand has been predicted to exceed 800 tons in 2020 (increasing from 400 tons in 2011)[54] and over the next 25 years, dysprosium demand will increase by 2600%[50].

In the past, MLCC electrodes were fabricated with precious metal electrodes such as platinum or silver-palladium alloys. In the late 1990's however, cost reductions were made by using base metal nickel internal electrodes[12, 13]. In a typical MLCC design, the internal electrode is made from nickel paste which is alternated between the dielectric layers. Copper paste is then applied as an external electrode, followed by an electroplated thermal barrier of nickel and finally an electroplated tin layer to improve the solderability[55]. This design allows for the minimum space to be used whilst achieving the maximum capacitance from a thin dielectric[56].

There are two different types of TECs; wet and solid. Solid TECs are prepared from tantalum powder which is pressed into an anode using a binder (the tantalum leads are inserted into the pellet at this time). Once sintered a dielectric layer of tantalum oxide (Ta_2O_5), up to 1.1µm thick, is formed on the surface of the anode through electrolysis[15]. A layer of manganese dioxide (MnO_2) is then formed around the Ta/Ta_2O_5 and acts as the cathode[57]. Graphite is layered between the MnO_2 and silver paste to avoid reduction of the MnO_2 and oxidation of the silver. The graphite and silver combination eliminates the use of tantalum foil which reduces cost, weight and improves performance[10]. Finally, epoxy resin is used to encapsulate the capacitor and tin is used as a termination[58]. In a 'wet' capacitor the tantalum anode is held in a liquid electrolyte[57]. At low temperatures, the wet TEC exhibits an increase in resistance but the electrolyte may permeate into the seal which dries out the capacitor over time and reduces its lifespan[59].

2.4 Capacitor recycling

A review of literature has found very little information on the specific disposal or recycling routes of capacitors but some work has been conducted on the disassembly of electrical components (which includes capacitors) from waste PCBs (WPCBs). Chen et al.[60] have documented that unregulated recyclers in developing regions such as Africa and Asia use handmade tools to disassemble electrical components by heating a WPCB on a coal-heated plate in order to melt the solder. This causes severe pollution to the environment and exposure to toxic chemicals for those involved in the work. In 2011, China banned these activities and now, along with India, employ semi-automatic techniques which involve infrared heaters and hot fluids like diesel to melt the solder[60]. Wang and Xu[6] discussed dismantling electrical components from WPCBs by damaging the joints between the electrical components and the WPCB, dissolving the solder by a chemical reaction or using heat and then applying an external force to free the electrical component from the WPBC. Wang and Xu[6] further reported that following disassembly, electrical components are recycled for precious metal recovery using mainly the hydrometallurgy technique (chemical leaching in combination with complexing agents, for example oxalic acid)[60, 61], although there is currently is no mature technique for this procedure. Pyrometallurgy can also be used to recover electrical components which involves heating WEEE to temperatures above 1000°C to recover the required metals, thus leading to high energy consumption and hazardous gas emissions. Rocchetti et al.[9] developed a portable system called HydroWEEE to recover base and precious metals from WEEE residues. This process involves hydrometallurgical treatment of WPCB granulate, i.e. with the electrical components intact. In Europe, the output from this process is currently landfilled or treated by pyrometallurgy plants. In this case, the copper extraction phase, performed with sulphuric acid and hydrogen peroxide, resulted in the highest impact across all of the reported categories[6, 9].

Although at the time of their disposal, most electrical components have only reached around 5% of their designed lifespan, reuse is frowned upon due to the possible instability of the component following refurbishment and also commercial sensitivities[60]. When WEEE is not collected for disposal or recycling, it is often stockpiled by consumers, again reducing the amount of reuse and recycling of finite materials[62].

A review of literature has found no information on the recovery of Dy, Ho or other rare earth elements from MLCCs or specifically BT. As rare earth elements have been identified as critical materials by a number of different organisations, this may become a crucial line of investigation in the near future[63]. The HydroWEEE system precipitates yttrium (a rare-earth metal) using oxalic acid but this is noted to be of high environmental impact due to the manufacturing process of oxalic acid and Rocchetti et al.[9] suggested that future research addresses the requirement for a new agent or process. Investigations into the recycling of tantalum from capacitors include Mineta and Okabe[58] who describe a two-step process involving initially tantalum recovery as an oxide followed by metallothermic reduction and leaching to collect metallic tantalum, yielding 99% purity Ta. Von Brisinski et al.[64] used a $AlCl_3$ based ionic liquid to isolate the Ta, dissolve the other metals (e.g. manganese, tin and silver) and thereby recover a number of materials.

2.5 Life Cycle Assessment of functional materials

The LCAs of functional materials and devices is evolving. For instance, Nease and Adams[65] used the process LCA methodology to compare the environmental impacts of bulk scale solid oxide fuel sell power plants fueled by gasified coal, with those fueled by a combination of pulverised coal and integrated gasification. Their results highlighted that with carbon capture enabled, a coal-fed solid oxide fuel cell plant can have a lower impact than a modern natural gas plant[65]. Strazza et al.[66] carried out the LCA of solid oxide fuel cells used as auxiliary power systems on boats. The work highlighted the fuel production phase as the highest impact within the life cycle and recommended the use of bio-methanol as a fuel to reduce this impact.

A study by Ibn-Mohammed et al.[67] on the comparative hybrid LCA of potassium sodium niobate and lead zirconate titanate outlined the increased impact of niobium mining which outweighs the impact of lead across five toxicology impact catergories including human toxicology. LCA of Perovskite solar cells (PSCs) have been investigated by several leading authors. Zhang et al.[68] and Ibn- Mohammed et al.[69], both used the hybrid LCA methodology to examine the environmental viability of PSCs. Ibn-Mohammed et al.[69] show that solar cells based on perovskite structures offer a more environmentally friendly option and ultra-low energy payback period when compared with existing photo voltaic cells, while Zhang et al.[68] discuss the merits of substituting silver or aluminium for gold in the production process due to the high impact of gold on the overall lifecycle. Ahmed *et al.*[70], considered LCA and technoeconomic analysis of triboelectric nanogenerators (TENGs), where it was highlighted that future research into TENGs should focus on improving system performance, material optimization and more importantly improving their lifespan to realize their full potential.

Despite the interest in other functional materials and devices with respect to LCA, and their importance in modern technology, there is a dearth of LCA work on electronic passive components such as capacitors. As highlighted in section 1.2, only one LCA study on MLCC was found in the extant literature and the environmental profile information contained therein were limited in scope due to the commercial sensitivity of the results[65]. Nevertheless, this study[20] adopted the Eco indicator 95 method to determine the environmental impact of MLCCs, surface mounted resistors and conventional resistors. A detailed bill of materials (BOM) is not published due to confidentiality but it is noted that a silver-palladium alloy is used as the internal electrodes. The LCA work showed that electricity consumptions and the ceramic powder are the highest contributors to the overall environmental impact. Primary data was used with the addition of public data and literature used to fill gaps where required, this enabled the authors to include packaging and waste in the assessment. It submitted that only 20% of purchased material was used in the final MLCC product[20].

3. Fabrication route for laboratory-based MLCCs and TECs

In this section, simplistic procedures for fabricating both the MLCCs and TECs are presented (Figure 4). Although simplified, the procedures broadly follow those anticipated in industry but where information is not available since it is commercially sensitive, laboratory based data is substituted[44].

3.1 Fabrication route for MLCCs

The MLCC production process can be broken down into five basic steps: i) ceramic production; ii) electrode printing; iii) layering; vi) heat treatment and v) termination. As shown in Figure 4 (right hand side), the process begins with BT powder preparation in which a solid state reaction between barium carbonate and titanium dioxide at 900°C for 6 hours yields BT[48]. This material is then milled, dried and mixed with the appropriate solvents and binders. The tape casting process then produces green BT layers, typically 2-5µm in thickness[12]. Nickel electrodes are then printed on to the ceramic tape, which are then stacked, the green body cold isostatically pressed and then cut to size[46, 71, 72]. Binder burn out takes place at 400-600°C for 2 hours followed by sintering at approximately 11-1200°C for 6 hours. The first termination layer is copper, this makes contact with the internal electrode; in order to protect the component during soldering, a nickel thermal barrier layer is electroplated on to the copper termination; finally, tin is electroplated on to the nickel termination to improve solderability[46]. The total time required to produce one MLCC in a laboratory environment is almost 76 hours.

3.2 Fabrication route for TECs

Production of a TEC (Figure 4, left hand side) in a laboratory takes approximately 17 hours and begins with the pressing of ground tantalum powder into a pellet (at which time the Ta leads are inserted into the pellet) followed by sintering at 1700°C for 30 minutes under a vacuum. A dielectric layer of Ta_2O_5, up to 1.1µm thick, is formed on the surface of the anode through electrolysis; the pellet is immersed in 0.1% phosphoric acid electrolyte solution[10]. The manganese dioxide (MnO_2) cathode is formed through pyrolysis of liquid manganese nitrate, the Ta/Ta_2O_5 pellet is immersed in manganese nitrate until 100% coverage of the dielectric layer is reached and then water is evaporated[10, 73]. Electrical contact is made with the MnO_2 by the application of a carbon layer which also protects the layers underneath from thermal and mechanical shock that may be caused by future processing[73]. A conductive silver layer is then applied, followed by epoxy resin and tin for soldering[10, 58, 73].

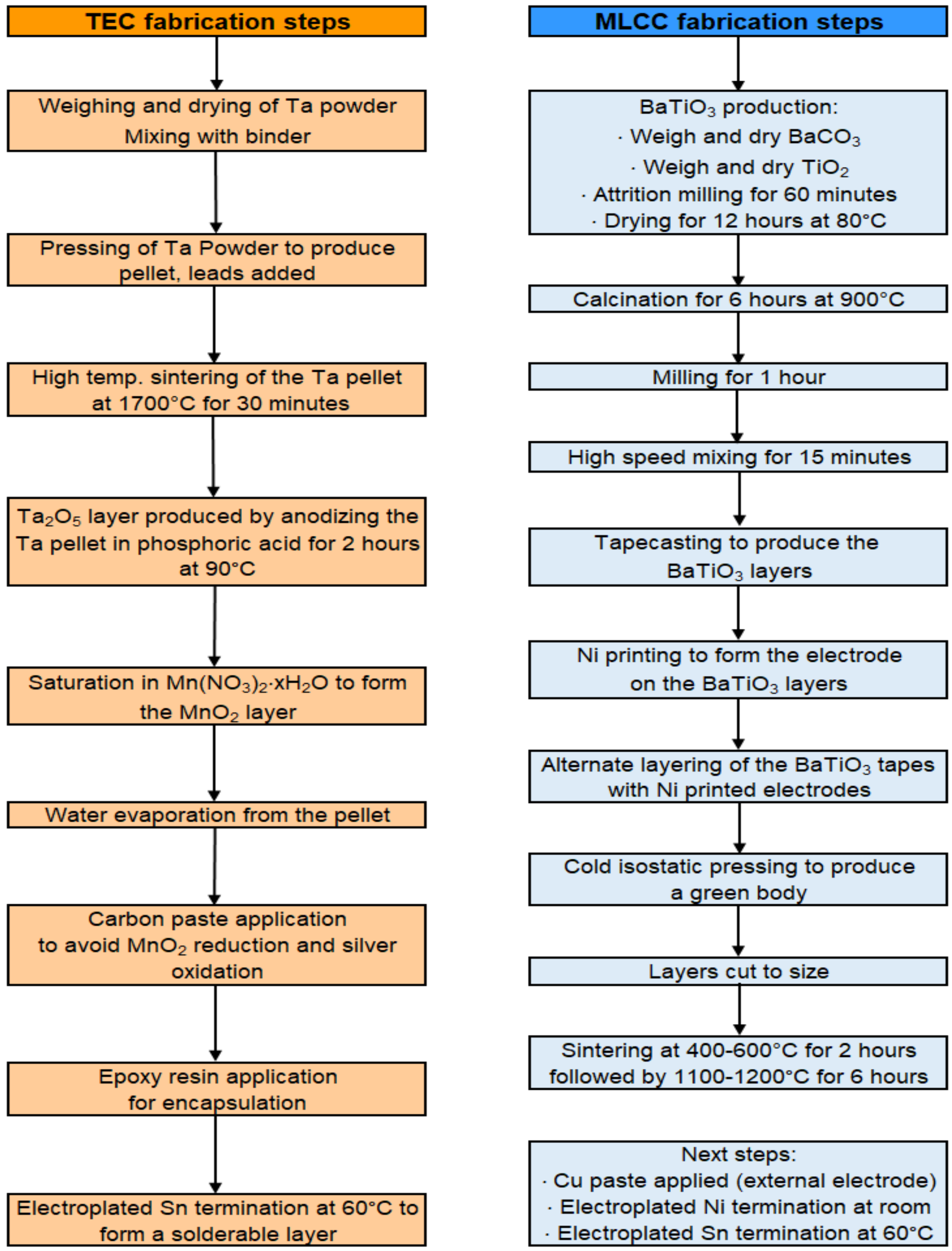


Figure 4: Fabrication route of MLCC and TEC volumetric efficiency capacitors. The procedures broadly follow those anticipated in industry but where information is not available since it is commercially sensitive, laboratory based data is substituted which approximates that used in industry[64].

4. Research Methodology

In this section, a detailed methodological framework for the comparative environmental profile evaluation of TECs vs MLCCs is presented based on the systems boundary depicted in Figure 5.

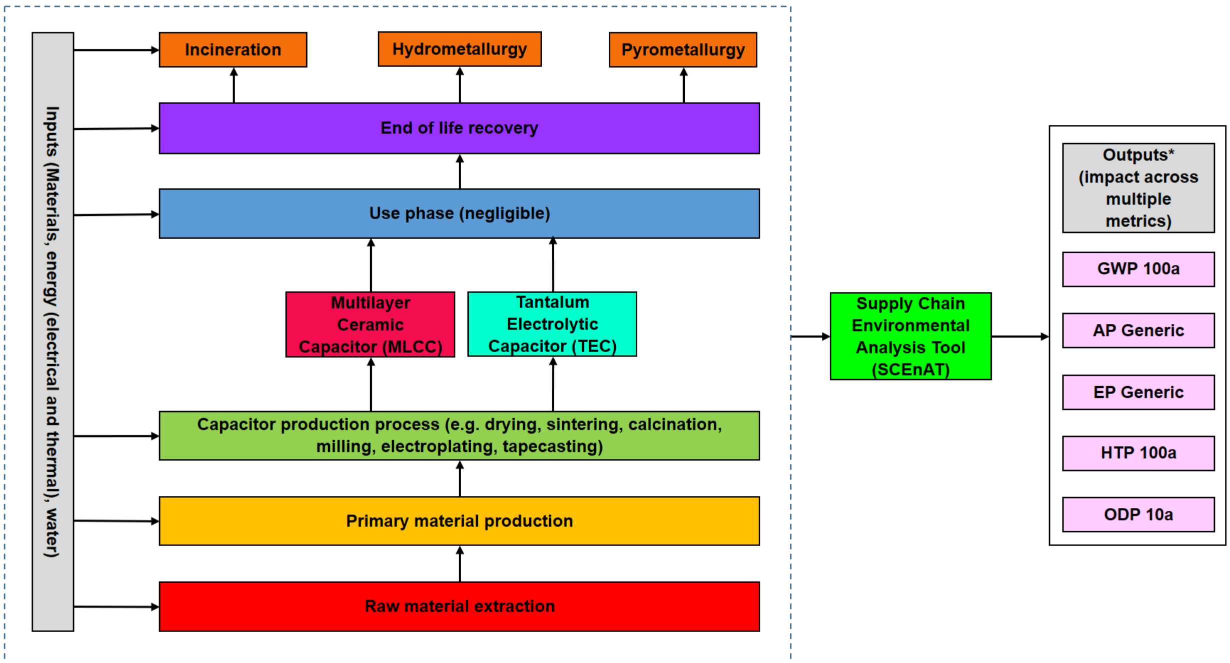


Figure 5: LCA system boundary, capturing the materials and energy flows associated with the fabrication processes of both TEC and MLCC. For detailed life cycle inventory upon which the system boundary is based, see Supplementary Material.

4.1 Life Cycle Assessment Framework

Life Cycle Assessment (LCA) is a structured framework for the assessment and estimation of the environmental impacts associated with a material, product or service[74]. These environmental impacts include (but are not limited to) climate change, acidification eutrophication, ozone depletion, water use and human toxicity[75]. Guinee et al.[75] discuss the past, present and future trends of LCA in their review of the subject. Since the turn of the century LCA has been put into practice through implementation in European Policy and throughout the world. Currently, and in the future, LCA will account for all three dimensions of sustainability - the environment, society and economics[75]. BS EN ISO 14040:2006 outlines four phases in an LCA study: goal and scope definition;

inventory analysis; impact assessment and interpretation[76-78]. The overall LCA consist of the following steps: i) identification of the raw material requirements, production and fabrication processes and energy requirements of both TECs and MLCCs; ii) establish systems boundary to consider and determination of a functional unit; iii) development and construction of the life cycle inventory and iv) overall impact assessment and environmental profile evaluations across multiple sustainability metrics.

In this work, the functional unit is 1 kg for each MLCCs and TECs with capacitance of 1μF and all of the inventories generated are converted by aligning them to conform to the functional unit. The motivation for this work pertains to climate change challenges due to greenhouse gas emissions. However, Ibn-Mohammed et al.[67] demonstrate the importance of considering other sustainability indicators which allows for detailed trade-off analysis. Accordingly, seventeen (17) environmental impacts were chosen from the Ecoinvent database to compare the impact of the individual components of each type of capacitor. Examples include the global warming potential (GWP 100a), acidification potential, eutrophication potential, human toxicity potential, land use and oxygen depletion potential. The primary energy demand is quantified using the cumulative energy demand impact factor. The Eco indicator 99 impacts are presented as complementary to the CML2001 impacts to allow for further assessment. A list of the remaining impacts can be found in Table S9 of the supplementary material. All the spectrum of metrics considered are in line with the Indicators of Sustainable Development identified by the United Nations Commission's Sustainable Development Framework[79]. More importantly, the chosen impact categories must be relevant to the requirements of the LCA[80]. At the moment, there is no universal list of impact categories that exist but LCA professionals choose categories based on the scope of the study[67].

All material use is assumed to be virgin material. While the MLCC industrial process is reported to have only 20% efficiency (wastes include ceramics, ancillaries, pastes and plating solutions)[20], the laboratory process is much more efficient and therefore only plating solution waste and passivation solutions (TEC production) are applicable, all of which are utilized to saturation.

4.1.1 Life cycle impact assessment modelling

Process-based LCA and Environmental Input-Output (EIO) LCA are the two main LCA techniques for computing environmental burden of a product or activity[67, 81, 82]. Process-based LCA works by establishing a system boundary based on the scope of the study, accounting for individual emissions contributions within the system[67]. This is achieved by multiplying the quantity of a material or a unit process (Q) by the emissions intensity (E_i) of the materials and processes as illustrated in Equation 1:

$$Process\ LCA = \sum_{i=1}^{n} Q_{p(i)} \times E_{p(i)} \tag{1}$$

However, LCA study based purely on process-based approach suffers from some degree of incompleteness due to systems boundary truncation[83]. To account for such truncation in boundary, LCA practitioners have leveraged economic input–output information (known as economic input–output (EIO) LCA) to quantify environmental life cycle impacts across economic sectors based on Equation 2:

$$EIO\ LCA = E_{io}.(I - A)^{-1}.y \tag{2}$$

where: $E_{io}.(I - A)^{-1}$ is the total (direct and indirect) emissions intensities of each industry required to produce the final demand product.

The integration of both process-based LCA with EIO LCA[27, 84-86] into a consistent framework based on hybrid LCA[67, 81, 82, 87] can provide much more robust results by expanding the system boundary and complies with ISO standards[88]. As this hybrid LCA process assesses the complete supply chain, providing full visibility, it is important to apply the methodology to cases such as those presented in this paper. A decision support tool known as the Supply Chain Environmental Assessment Tool (SCEnAT) developed by Koh et al.[89] integrates both process-LCA and EIO LCA and is employed to compute the environmental profile of the capacitors under consideration. The framework of the tool is based on five steps namely: supply chain mapping, carbon calculation, low carbon interventions, supply chain performance evaluation and informed decision making. This tool has been successfully implemented with a number of companies yielding environmental improvements within their supply chains[89, 90]. The results of each Hybrid LCA are compared to determine which capacitor poses the highest environmental impact, consequently providing information to be used at the design phase of electronic devices.

In this work, three end of life methods including incineration, hydrometallurgy and pyrometallurgy were considered. The Ecoinvent database was adopted to determine the 'consumer to grave/cradle' impact of 1kg of MLCCs and 1kg of TECs, yielding a number of capacitors each with 1µF capacitance; the 'treatment of used capacitors, to hazardous waste incineration' dataset was utilised in conjunction with the 'treatment of average incineration residue' dataset in order to take into account the impact of the waste arising from the incineration process. The hydrometallurgy and pyrometallurgy metal recovery routes were mapped in the SCEnAT decision support tool to determine which of the three (currently) feasible processing routes lead to the lowest environmental impact based on the categories chosen. Due to the limitation of data availability in the Ecoinvent database, the datasets corresponding to hydrometallurgical and pyrometallurgical treatment of a lithium-ion battery were used to represent the capacitors in question. Rochetti et al.[9] describe the recycling of Li-ion accumulators by hydrometallurgy as a similar process to that of PCBs using sulphuric acid leaching, neutralisation, metal recovery and waste water treatment. Bernardes et al.[91] describe the pyrometallurgical process of Li-ion battery recycling as utilising higher temperature of that for other electronic components. Following both hydro- and pyrometallurgical metal recovery there is a residue that is untreatable and therefore must be sent to landfill, consequently the 'treatment of average incineration residue' dataset has been used to represent this impact[92].

4.1.2 Choice of functional unit

For any LCA work, the overall aim is always to gain an understanding of the environmental profile of a given system of processes that together delivers a defined function. Accordingly, the most essential quantity that defines the scope of an LCA study is termed the functional unit[77]. This specifically defines the type and size of the product (or, more generally, some activity or even service), the life cycle of which is being assessed by quantitatively describing the function it delivers. In this work, given that the LCA of two types of capacitors are under consideration, the functional unit is therefore selected on the basis of the capacitance in microfarads (µF) of the capacitors. However, in practice, due to the tiny nature of capacitors, they are fabricated in batches. As such, the functional unit adopted in this work is on the basis of how many capacitors with respective capacitance can be produced using 1 kg of the entire material inventory for each MLCCs and TECs. Following on from this, for the MLCCs, 1 kg of the entire material inventory yielded 670,630 capacitors, each with a capacitance of 1 µF, and for the TECs, the total number of capacitors produced is 33,697. The schematics for each of the capacitors are illustrated in Figures 6 and 7 below.

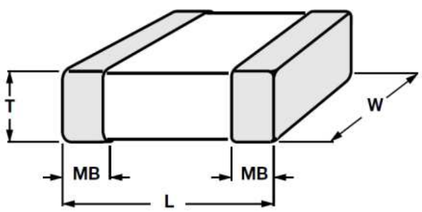


Figure 6: Schematic of a MLCC used as a basis of the functional unit[93].

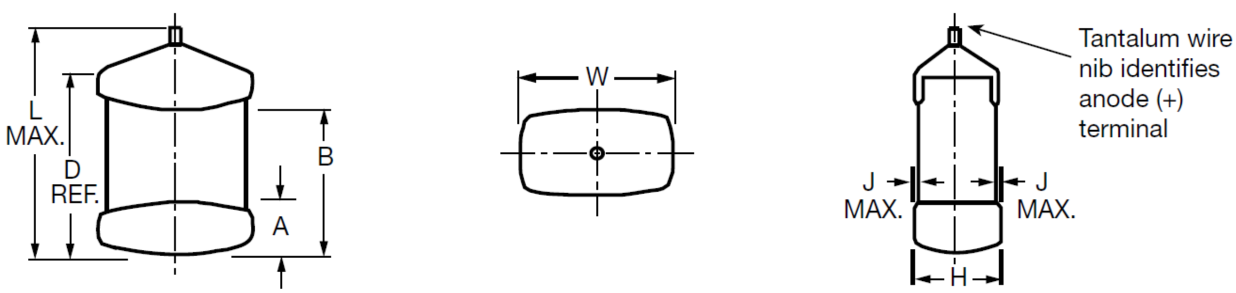


Figure 7: Schematic of a TEC used as a basis of the functional unit[94].

Figure 6 shows a schematic of the MLCC used as a bases of the functional unit; in this case, L is 1mm, W is 0.5mm, T is 0.5mm and MB is 0.25mm[93]. Figure 7 shows a schematic of the TEC used as a basis of the functional unit; in this case, L_{max} is 2.2mm, W is 1.1mm, H is 1.1mm, A is 0.4mm, B is 1.07mm, Dref is 1.6mm and J_{max} is 0.1mm[94]. The thickness of the MLCC end terminations (tin, nickel and copper) and the internal electrode thickness were given by Lee et al. for MLCCs[95]; such data was not available for TECs and therefore assumptions were made using the Lee et al. data and applied to the TEC termination for coatings and electrode thicknesses.

4.2 Data sources

Due to the complexity of supply chains, data collection processes involved in any LCA study can be intensive depending on the scope and the nature of the products or activity under consideration. It is best to use primary data as much as possible. In instances where primary data are not available, the life cycle inventory (LCI) can be augmented using secondary sources[96]. In this contribution, most of the primary data were derived from the laboratory. The Ecoinvent database[21] was used to provide the background data in the form of environmental impact categories. BS EN ISO 14040 standard defines "selection of impact categories and classification" and the impacts chosen should be of relevance to the study[76]. Dreyer et al.[80] compare the methodologies of EDIP97, CML2001 and Eco-indicator 99. CML2001 and EDIP97 represent impacts at the midpoint, i.e. somewhere between the source and receptor, whereas Eco-indicator 99 represents impacts at the end-point, i.e. the receptor. The land use impact category is represented in CML2001, but not in EDIP97, while EDIP97 models a waste category unlike CML2001. Due to the difference in modelling, it is not possible to directly compare all three of the methodologies. In this work, the analysis provided were based on emissions intensity data derived from CML200l impact categories as detailed in Ecoinvent database.[21]

4.3.1 Construction of life cycle inventory for the LCA

The structure of a MLCC and a TEC are outlined in section 2.1; the BOM[3, 10, 15, 57, 58, 97-100] used to determine the impact of 1kg of each type of capacitor are outlined in Tables S1 and S2 of the supplementary material. It is important to note that additional minor components are required in the manufacture of a capacitor, such as binders. The production method for each type of capacitor is outlined within section 3 and presented in more detail in Tables S3 and S4 of the supplementary material. The information used to construct the LCI was derived from well-established data from within the literature, laboratory process based engineering knowledge, study assumptions and upstream emissions data from the Ecoinvent database[21]. For materials whose emissions intensity data were unavailable, data were derived on the basis of stoichiometric reactions based on previously published guidelines and substitution based on chemical characteristics or functional similarities[67, 101]. The EIO dataset were based on the supply and use table for 2008 which is embedded within the overall framework of the SCEnAT modelling tool.

The MLCC dimensions were given by Vishay for the X7R 0402 MLCC[93]; TEC dimensions were given by Vishay for the 595D case cade T TEC[94]. The thickness of the MLCC end terminations (tin, nickel and copper) and the internal electrode thickness were given by Lee et al. for MLCCs[95]; such data was not available for TECs and therefore assumptions were made using the Lee et al. data and applied to the TEC termination for coatings and electrode thicknesses.

Given that all manufacturing procedures are conducted using electrical equipment in the laboratory, the electrical energy consumption (kWh) is calculated by multiplying the electrical power (W) of the specified device as stated by the manufacturer by the time (sec). To account for thermal energy requirements of the manufacturing processes, the required energy (Q) is calculated by multiplying the specific heat capacity of the material heated (J/kg·K), mass of material heated in the process (kg) and temperature difference (K or °C). A capacitor is an energy storage device and does not use energy. Therefore, the use phase of a capacitor must be considered by its dielectric loss (tan δ), which refers to the reduction in power between the applied *ac* voltage and current[33, 102]. BT has been found to have a tan δ of 0.012, i.e. 1.2% of the energy stored[103]. Consequently, the use phase was calculated to be negligible (see supplementary material) and therefore was omitted for the scope of this investigation[12]. As stated in section 2.4, capacitors have usually only reached around 5% of their designed lifespan when they reach the disposal phase. Consequently, the cycle life of each impact need not be considered in the overall comparative analysis of the two capacitors.

5. Results and Discussion

5.1 Primary energy consumption

Figures 8 and 9 show the overall distribution of the primary energy consumption for the fabrication of a laboratory-based MLCC and TEC. Specifically, Figures 8a and 9a indicate the total primary energy consumption, including materials embedded (i.e. embodied energy in natural resources attributed to extraction)[67], thermal and electrical energy relating to each of the manufacturing process; MLCC totalling 5567.65 MJ-eq/kg and TEC totalling 6862.29 MJ-eq/kg. As shown, materials embedded constitute the highest impact from primary energy demand for TECs, while electrical energy is the highest contributor for MLCCs. Figures 8b and 9b show the percentage contributions of each of the process steps regarding the thermal energy consumption. A breakdown of the material embedded in MLCC fabrication (Figure 8c) shows that the use of nickel paste is the outweighing component, contributing over 49% of the material impact category. Figure 9c shows that roughly 97% of the material embedded energy in TEC fabrication is attributed to the use of tantalum. The percentage contributions of each of the process steps with regards to electrical energy are shown in Figures 8d and 9d. The drying process (Figure 8d) constitutes 62% of the entire electrical energy consumption for MLCC fabrication. “Others” in Figure 8d represent those inputs lower than 1%, namely: weighing, high speed mixing, cold isostatic pressing and aging of the paste. In the case of TECs, the sintering process (Figure 9d) constitutes the largest consumer of electrical energy, representing about 64%. This suggests that drying and sintering processes are the main hotspot for both MLCC and TEC for which mitigation strategies should be targeted. “Others” in Figure 9c represents those inputs lower than 1%, namely: graphite paste, silver paste, epoxy resin and silver termination. In Figure 9d, “Others” represents weighing, pressing and water evaporation (again, those inputs under 1%).

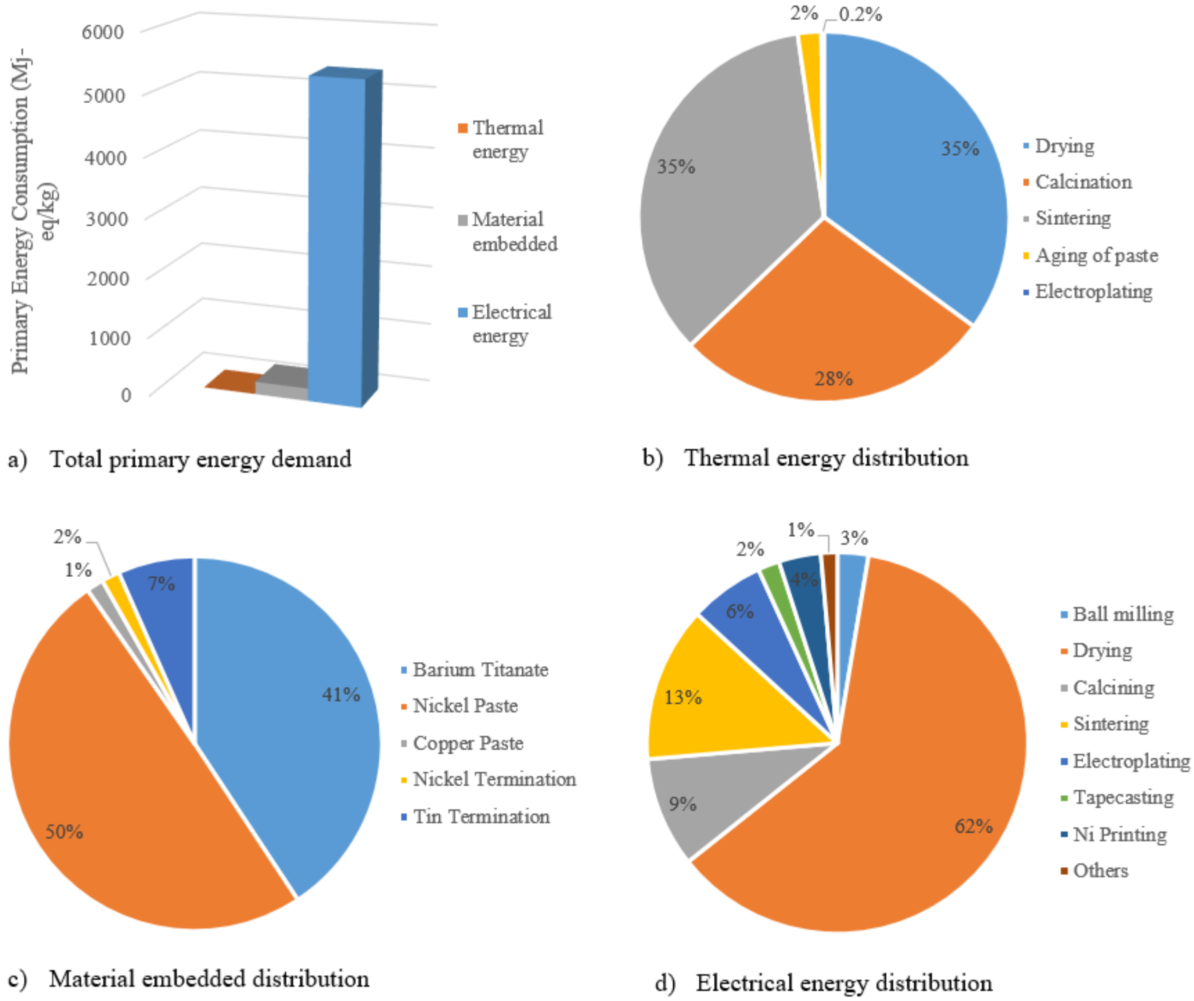


Figure 8. Distribution of the primary energy consumption for the fabrication of an MLCC (a) Total primary energy consumption including thermal and electrical energy and materials embedded all expressed in MJ kg-1. (b-d) indicate the percentage contributions of each process or material relative to (a). For a detailed breakdown of the supply chain map, see Figures S1 and S2 of the supplementary material.

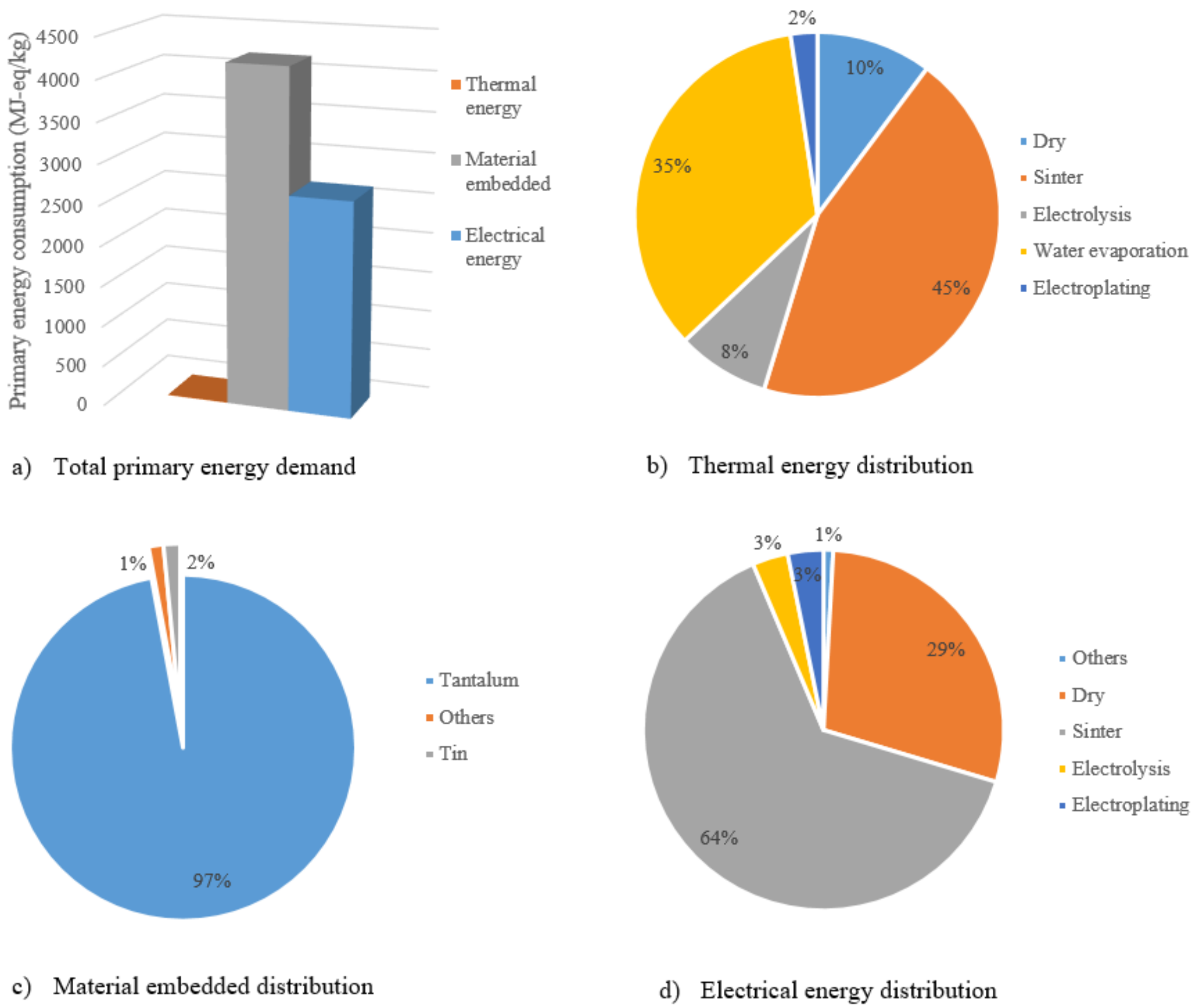


Figure 9. Distribution of the primary energy consumption for the fabrication of a TEC (a) total primary energy consumption including thermal and electrical energy and materials embedded all expressed in MJ kg^{-1}. (b-d) indicate the percentage contributions of each process or material relative to (a). For a detailed breakdown of the supply chain map, see Figures S1 and S2 of the supplementary material.

Figure 8a shows that electricity usage is the carbon hotspot in the manufacture of a MLCC, in agreement with the results presented by Philips[20]. High electricity use is required in the manufacturing stage due to the length of time required to complete the drying, calcining and sintering production phases of BT. The overall impact is likely to be reduced in industry due to larger, more efficient machinery with high batch throughput[104]. As identified by Ibn-Mohammed et al.[67], optimised sintering approaches such as the use of sintering aids and low temperature processing technology can contribute to the overall reduction in thermal and electrical energy demand for fabrication of functional materials. Cold sintering – a process based on the addition of small amounts of water to aid the key transport processes that densify the materials for device development has also been touted as a means for lowering sintering temperatures[105-107]. On the other hand, about 61% of the primary energy demand for the fabrication of a TEC is caused by the materials embedded (Figure 9a) for which the use of tantalum pellet (including the tantalum leads) constitute 97% of the overall impact (Figure 9c). Therefore, raw material extraction is the major source of environmental impact. Ta is almost always found with niobium in nature due to their similar chemical naturestics[108]. Its extraction is very energy intensive and includes activities such as blasting, crushing, smelting and separation[24, 67].

5.2 Component level analysis

Figures 10 and 11 show the component level analysis of the environmental impacts of MLCC and TEC fabrication processes respectively. This was undertaken to identify their influential components and materials across a number of sustainability metrics which are normalised, ensuring that the absolute indicator of each category of impact is 100%. Given that the impact from electricity and natural gas use are illustrated in Figures 8b and d and Figures 9b and d, they have been omitted from Figures 10 and 11 to highlight the most influential materials responsible for the overall environmental impact of both capacitors.

Figure 10 shows that the use of nickel paste has the highest percentage impact for climate change (51%), acidification (75%), eutrophication (58%), high NO_x POCP (71%), land use (65%), fresh water aquatic ecotoxicity (69%), fresh water sediment ecotoxicity (69%), human toxicity (52%), marine aquatic ecotoxicity (68%), marine sediment ecotoxicity (69%) and cumulative energy demand (50%). Ni is a vital metal in modern infrastructure and technology, with a wide range of applications[109]. A detailed analysis of local issues pertaining to the mining of Ni is provided by Mudd[109], where he submitted that although the environmental impact of Ni has improved across the years, its mining has resulted in serious historical local impacts including acid rain from SO_2 emissions, wetland acidification, soil contamination due to heavy metals, biodiversity loss (e.g. in fish populations). Nickel inhalation has been reported to lead to an increased risk of cancer in the lungs and noses of humans[110]. The remaining cases, i.e. ozone depleting (60%) and low NO_x POCP (75%), have the highest impact from the barium titanate component.

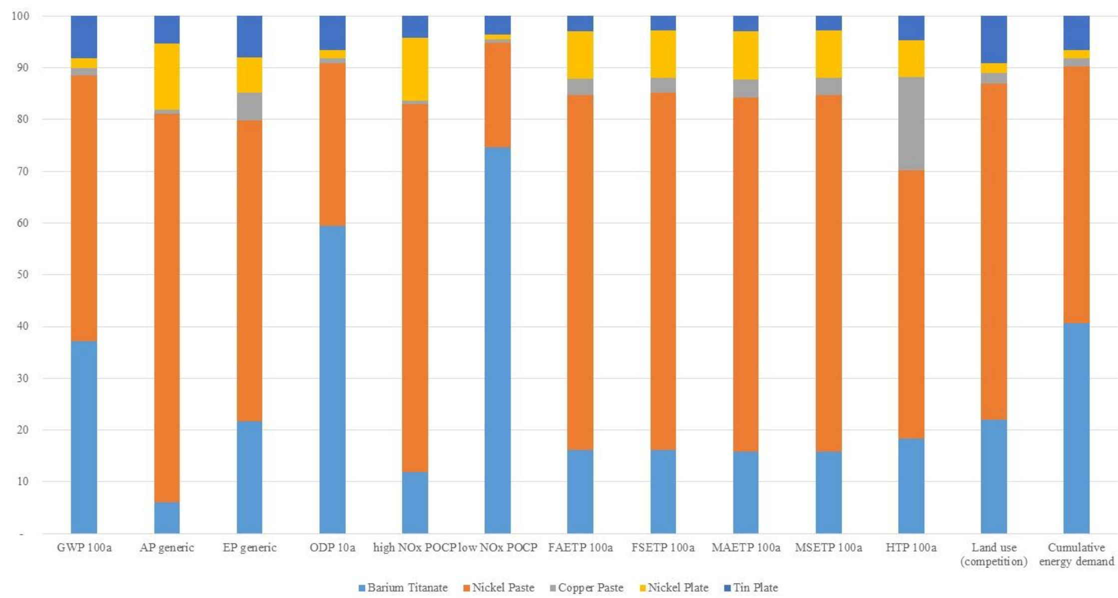


Figure 10: Percentage contribution of each MLCC manufacturing component of the environmental impacts investigated.

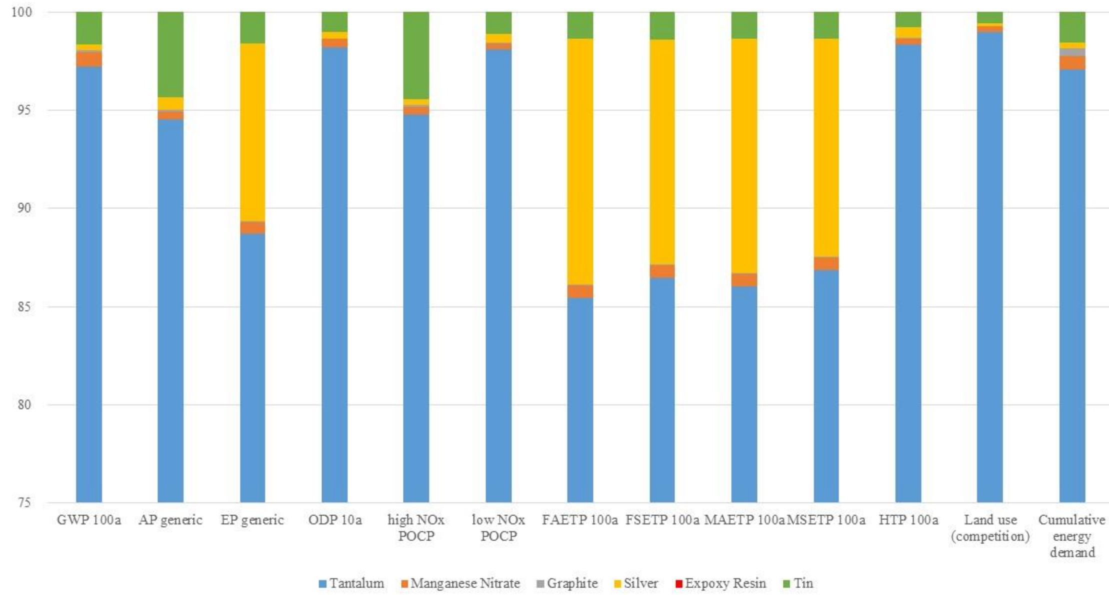


Figure 11: Percentage contribution of each TEC manufacturing component of the environmental impacts investigated. The y axis is shown from 75% to 100% to show the impact contribution from all materials.

As shown in Figure 11, the use of Ta in a TEC (pellet and lead) causes the highest impact of all of the components across all impact; climate change (97%), acidification (95%) eutrophication (89%), ozone depletion potential (98%), high NO_x POCP (95%), low NO_x POCP (98%), land use (99%), fresh water aquatic ecotoxicity (85%), fresh water sediment ecotoxicity (86%), human toxicity (98%), marine aquatic ecotoxicity (86%), marine sediment ecotoxicity (87%) and cumulative energy demand (97%). Given that the extraction mechanism of niobium is similar to that of tantalum since they are found together in nature, a number of approaches that can be adopted during their extraction to minimise overall impact are provided by Ibn-Mohammed et al.[24, 67]. The second largest contributor to the impact of TECs is the silver paste which is attributed to mineral extraction and the subsequent processes required to obtain the finished material[111].

Metal mining processes (for nickel and tantalum) are driven by the ore properties, tonnage, grade and depth. The most frequently used methods are surface or underground mining (or in combination). Of the two methods, underground mining requires more infrastructure and therefore leads to a higher environmental impact[111, 112]. Mudd[109] discuss that as mines go deeper to meet market needs, production and environmental costs increase. Large amounts of tantalum are extracted from the ground by artisanal and small scale mining (ASM). ASM, although sometimes formal, is often an informal activity conducted by small groups in developing countries. This type of extraction provides jobs and an income for millions of people but can lead to dumping of waste and effluent into rivers, deforestation, landscape destruction and land pollution (not an exhaustive list). These environmental impacts are usually caused by economic limitations and a lack of access to better techniques.[113, 114] ASM is likely to negatively impact the environmental indicators of tantalum.

Analysis was performed to determine how the manufacturing location would affect the electrical energy impact during MLCC manufacture. The Ecoinvent Great Britain data for ‘market for electricity, low voltage’ was compared to the same datasets for the United States, China, Japan and France. The highest impact was associated with China, the total impact for the electricity use in the manufacturing process of a MLCC was calculated to be 44.44 kg CO_2-eq; the lowest impact was associated with France, the total impact for the electricity use in the manufacturing process was calculated to be 4.35 kgCO_2-eq. This information shows that, of the countries compared, the most appropriate manufacturing location for energy consumption is France (see table S11 in the supplementary material).

5.3 Comparison of Environmental Profiles

Figure 12 highlights the key differences between the environmental profiles of TECs and MLCCs across a number of indicators. As already highlighted in the preceding sections, the overall environmental profile of TECs surpasses that of MLCCs across all impact categories except in the electrical energy. The thermal energy associated with MLCCs is 0.60 MJ-eq/kg compared to 4.39 MJ-eq/kg for that of TECs. This difference is due to the increased processing temperatures are material masses required in TEC production. 5352.77 MJ-eq/kg of electrical energy is associated with the production of MLCCs and 2665.82 MJ-eq/kg with that of TEC production. This difference can be attributed to the additional drying, milling, tapecasting, printing and calcining steps that are required for MLCC production but not for TEC production.

As highlighted in Section 5.2, nickel constitutes the highest environmental impact in the overall assessment of MLCCs but as indicated in Figure 12b, the toxicological footprints of TECs across all variants surpass that of MLCCs due to the use of tantalum. This is also the case in terms of the damage to ecosystem quality, resources and human health (Figure 12c). Figure 12d highlights the harmful effect of TECs on key economic sectors based on the upstream IO greenhouse gas emissions. The supply chain upstream impact of TECs is associated to its overall higher cost of production and the cost of the materials as compared to MLCCs. This assertion is particularly valid given that economic data such as cost of materials are converted into physical quantities (e.g. kg of material) in IO analysis. Accordingly, a higher conversion output will cause more upstream emissions across the supply chain of the material under consideration[67].

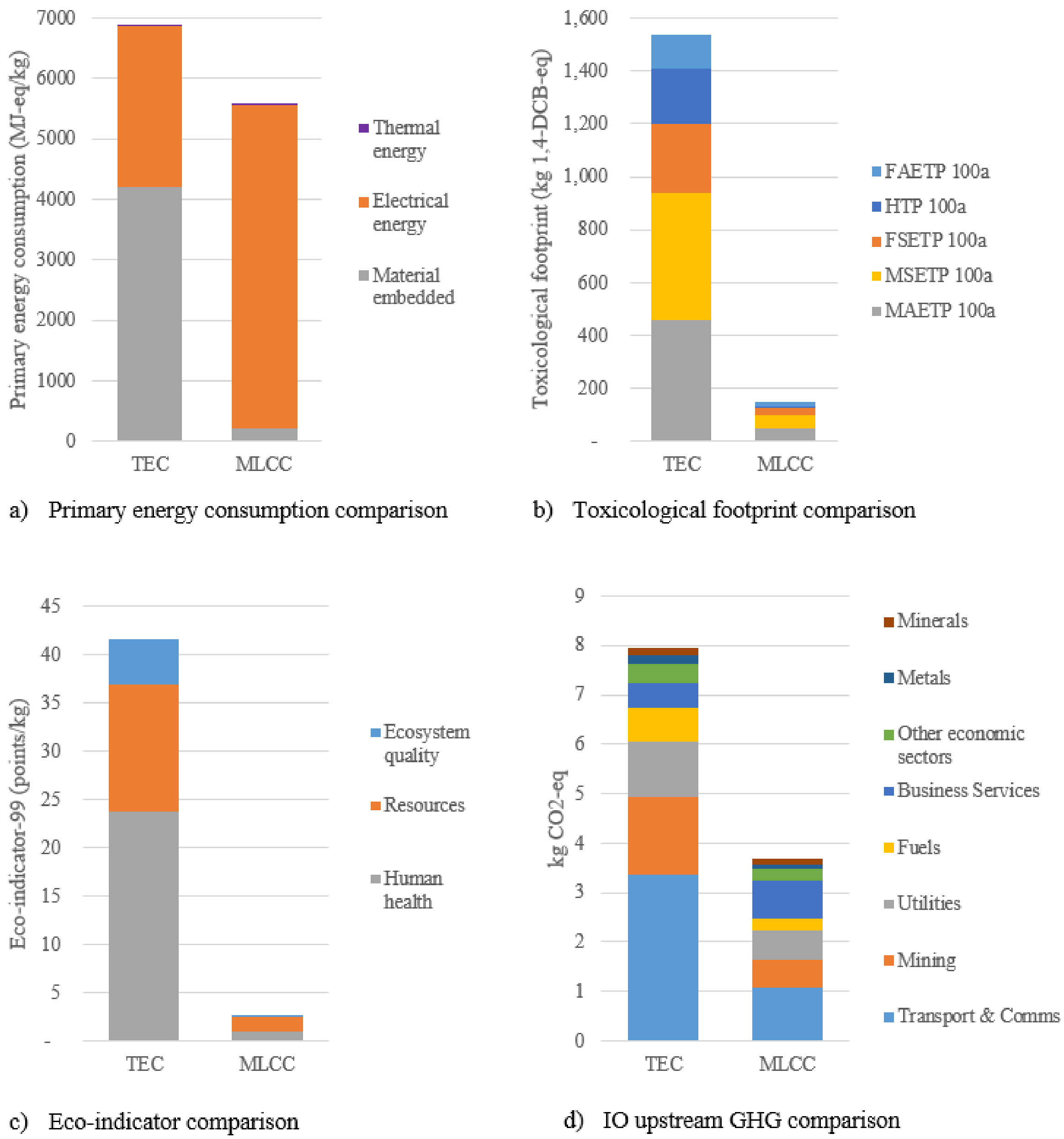


Figure 12 Comparison of TEC versus MLCC. a) Primary energy demand, b) toxicological footprint, c) eco-indicator 99 comparisons, d) IO upstream GHG comparison.

5.4 Impacts of end of life methods

The SCEnAT decision support tool was used to map the MLCC and TEC supply chains for cradle-to-gate, cradle to incineration, cradle to hydrometallurgy and cradle to pyrometallurgy and also to conduct the hybrid LCA carbon calculations and supply scenario analysis. Examples of the supply chain maps produced by SCEnAT can be found in Figures S1 and S2 of the supplementary material. The tool colour codes the supply chain to easily indicate the carbon hotspots; green represents <1% impact, yellow represents 1-5% impact, orange represents 5-10% impacts and red represents >10% impact. A comparison of the results provided for the MLCC and TEC supply chains by the SCEnAT tool is presented in Tables 2 and 3.

Table 2 SCEnAT Hybrid LCA calculations for a MLCC from Cradle to Gate, Incineration, Hydrometallurgy and Pyrometallurgy.

Parameter	**Units**	**Cradle-to-Gate**	**Cradle to Incineration**	**Cradle to Hydrometallurgy**	**Cradle to Pyrometallurgy**
Total Emissions	kg CO_2-eq	97.00	100.70	98.20	98.730
AP Generic	kg SO_2-eq	0.77	0.78	0.78	0.78
EP Generic	kg PO_4-eq	0.13	0.14	0.14	0.14
HTP 100a	kg 1,4-DCB-eq	22.87	23.92	23.78	25.13
Land Use	m^2a	10.14	10.39	10.22	10.25

Table 3 SCEnAT Hybrid LCA calculations for a TEC from Cradle to Gate, Incineration, Hydrometallurgy and Pyrometallurgy.

Parameter	**Units**	**Cradle-to-Gate**	**Cradle to Incineration**	**Cradle to Hydrometallurgy**	**Cradle to Pyrometallurgy**
Total Emissions	kg CO_2-eq	311.99	315.69	313.19	313.72
AP Generic	kg SO_2-eq	2.20	2.21	2.21	2.20
EP Generic	kg PO_4-eq	0.72	0.73	0.73	0.73
HTP 100a	kg 1,4-DCB-eq	214.54	215.58	215.45	216.80
Land Use	m2a	77.96	78.20	78.04	78.06

The SCEnAT analysis for each recovery method is compared. Of the three disposal/recycling routes investigated, the highest CO_2 emissions can be attributed to the incineration of a both MLCCs and TECs, as can the highest land use impact. The pyrometallurgical route leads to the highest human toxicity potential at 25.13 kg 1, 4-DCB-eq and 216.80 kg 1, 4-DCB-eq for MLCCs and TECs respectively. The pyrometallurgical process itself can include incineration, smelting, drossing, sintering, melting and high temperature reactions in the gas phase. The waste gases and flue dusts contain halogens, leading to dioxins that accumulate in the food chain and cause reproduction issues, immune system damage and cancer[115, 116].

The incineration process also causes the release of halogens[117] and the leaching of solutions used in the hydrometallurgy process can be toxic and corrosive[115]. The HTP 100a impact of hydrometallurgy may be lower than that of pyrometallurgy due to the possible wider spread impact of flue emissions compared to process fumes. That said, a leak or major spill of the leaching solutions would have a profound environmental impact on the ground and surrounding water systems. The acidification potential and eutrophication potential for incineration, hydrometallurgy and pyrometallurgy are equivalent. As acidification and eutrophication are caused by the combination of emitted gasses, such as SO_2, NO_x, HCl, dioxins and furans, it is possible to attribute the impact of incineration and pyrometallurgy to the atmospheric emissions produced during the combustion phase[115, 116]. With regards to hydrometallurgy, Rocchetti et al.[9] have deduced that the acidification potential and eutrophication potential impacts can be attributed to the recovery of yttrium using oxalic acid[9, 118, 119].

Min et al.[120] discuss the use of capacitors embedded into substrates to reduce the overall size of the substrate. Although this change may meet the market need for smaller components, it will add additional complexity to the disassembly phase of WEEE and is likely to lead to an increased loss of materials to landfill or energy recovery by incineration. This is an important example where the implementation of LCA in the design phase could lead to longer term environmental impact savings.

5.6 Limitations of the current work

Primary data derived from the laboratory was used for the comparative LCA of MLCCs and TECs based on the BOMs and the production processes. Other sources include publicly available data and literature. The absence of any further primary data is the main limitation of this study and therefore the reliance on the Ecoinvent database for the data required in the main scope of the investigation. Despite this inefficiency, this investigation is the most transparent published data available on the life cycle assessment of both a MLCC and a TEC. Any future work could include factors such as waste streams and packaging from primary sources in the scope of the LCA.

In an industrial setting, given the well-established manufacturing routes for both types of capacitors considered in this work, these components would be manufactured on a much larger scale than in a laboratory and therefore the high electricity used reported in the manufacturing process based on 1kg each for MLCC and TEC is likely to be lower because of the use of larger, more efficient machinery with high batch throughput[104]. Nevertheless, following discussion with engineers in Murata Manufacturing Co., Ltd. (a global leader in capacitors manufacturing) the environmental hotspots identified in this work are in line with processes adopted in an industrial setting. As identified by Ibn-Mohammed et al.[67], optimised sintering approaches such as the use of sintering aids and low temperature processing technology which are available in high tech industries can contribute to the overall reduction in thermal and electrical energy demand for fabrication of volumetric efficiency capacitors.

Hybrid LCA was adopted in this study to ensure the completeness of system boundary limitations of process-based LCA using EIO LCA data. However, the choice to include or exclude certain inventories from the EIO LCA data with the view to account for missing inputs whilst avoiding the double counting of inputs remains potentially subjective. Such missing inputs are chosen based on the discretion of the modeller and different results might be produced if another LCA modeller chooses different missing inputs. The use of Dy in a MLCC is diluted in the final output of the hybrid LCA by the high impact of electricity use. It is not highlighted as a hotspot in this study because of the low volumes in which it is used. Despite this, as the material is not currently recycled from these electronic components the earth's reserves are continually being depleted[60, 61].

6. Conclusion

In this work, a detailed cradle-to-grave analysis of the entire fabrication route of two representative capacitors is presented. No previous published work has been provided for either the LCA of a TEC or MLCC. In an era where environmentally-sensitive manufacturing procedures are rigorously monitored due to the increased global awareness of environmentally benign design and the strong relationship between global warming and CO_2 emissions, this is an important gap to fill. The electrical impact of a TEC is 2666 MJ-eq which is lower than that of a MLCC at 5353 MJ-eq, but the material embedded energy (i.e. the cumulative energy demand) of a TEC is 20 times that of a MLCC and therefore the overall primary energy demand of a TEC (6862 MJ-eq) is much higher than that of an MLCC (5567 MJ-eq). 97% of the global warming potential of TECs can be attributed to the use of tantalum for the pellet and the leads, due to the energy intensive nature of the extraction and purification process. Although the main drivers for replacement of TECs with MLCCs relate principally to cost, the need for the development of miniaturised versions of devices and longevity, this work further demonstrates that large environmental savings are an additional benefit. By replacing TECs with MLCCs in electrical components, a decrease in the environmental impact is achieved. Despite this improvement, it is also important for MLCC manufacturers to consider their designs and look to decrease further their environmental impacts.

A number of environmental hotspot mitigation strategies including the use of optimised sintering approaches such as the use of sintering aids and low temperature processing technology, cold sintering techniques and recommendations to minimise environmental impacts of metals used in the fabrication of capacitors is provided. Although capacitor recycling is not yet well established, this work shows that the hydrometallurgical recycling process leads to the lowest environmental impact when compared to incineration and pyrometallurgy. Research is required to fully exploit the material recovery possibilities of waste capacitors.

Any future research in this area would benefit from a primary data source. Furthermore, this work shows that it is imperative that research is carried out to reduce the environmental impact of MLCCs by reducing their reliance on rare earths and nickel internal electrodes. Overall, the methodological framework used in the current work should be useful for the LCA and environmental profile assessment of other emerging devices' architectures and technologies at the early stages before key design decisions are made.

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