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Life Cycle Assessment of aluminum alloys chips recycling through single and multi-step Friction Stir Consolidation processes☆

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ABSTRACT

Manufacturing scientists have to find new processes enabling energy and resource efficient circular economy strategies. Solid state recycling processes have proved to be environmentally friendly alternatives to recycle aluminum alloys process scraps like chips. In this paper the environmental characterization of a solid state recycling process named Friction Stir Consolidation (FSC) is presented. A full Life Cycle Assessment (LCA) comparative analysis is developed: the environmental performance of new and different variants of FSC processes and remelting based routes are quantified and compared to one another. Also, different scenarios are analysed to provide guidelines for the correct implementation of FSC based recycling processes. Results revealed that: 1/ FSC is an environmentally friendly solution although a correct industrial implementation is needed to reduce non-productive production steps, 2/ the transition towards a wider use of renewable energy sources would amplify the advantage of FSC with respect to conventional remelting route in terms of environmental sustainability.

1. Introduction

Production of just five key materials (aluminum, steel, cement, paper, and plastic) accounts for over half of all the greenhouse gas (GHG) emissions released by industry worldwide each year [1]. Primary aluminum production is the most energy and emissions intensive among the aforementioned materials [2]. Aluminum production industry is responsible for about 3 % of the world's 9.4 Gt direct industrial CO_2 emissions in 2021 [3]. Since 1971, the global demand for aluminum has increased by nearly six times; although during the Covid-19 pandemic the aluminum production fell flat, it has since started growing quickly once again and global demand is likely to continue growing in response to increasing global population and GDP [3]. The main approach for reducing the primary production and decoupling the resource depletion from the economic growth is the implementation of circular economy strategies [4]. Longer life, repair, product upgrades, modularity, remanufacturing, component reuse and recycling are some of the strategies to put in place to reduce the environmental impact of raw material production [5].

Manufacturing processes play a crucial role in putting in place the aforementioned strategies. Manufacturing scientists have to tackle such challenge and have to make a research effort addressed to find proper manufacturing processes enabling Circular Economy (CE) strategies. Overall, CE oriented manufacturing operations rise new challenges at different levels: form process unit and manufacturing systems [6] up to supply chain levels [7].

Moreover, the energy and resource efficiency of the CE processes needs always to be guaranteed and minimized both at machine level [8] and at manufacturing system level [9]. What is more, the impact of the manufactured semifinished/finished products should be characterized by a wider perspective including alle the factors of influence (materials, consumables, use phase impact, etc.) in the environmental impact analysis [10].

As far as metals are regarded, recycling is still the most used Circular Economy strategy. Nevertheless, it is worth remarking that for aluminum alloys the conventional recycling route, based on remelting,

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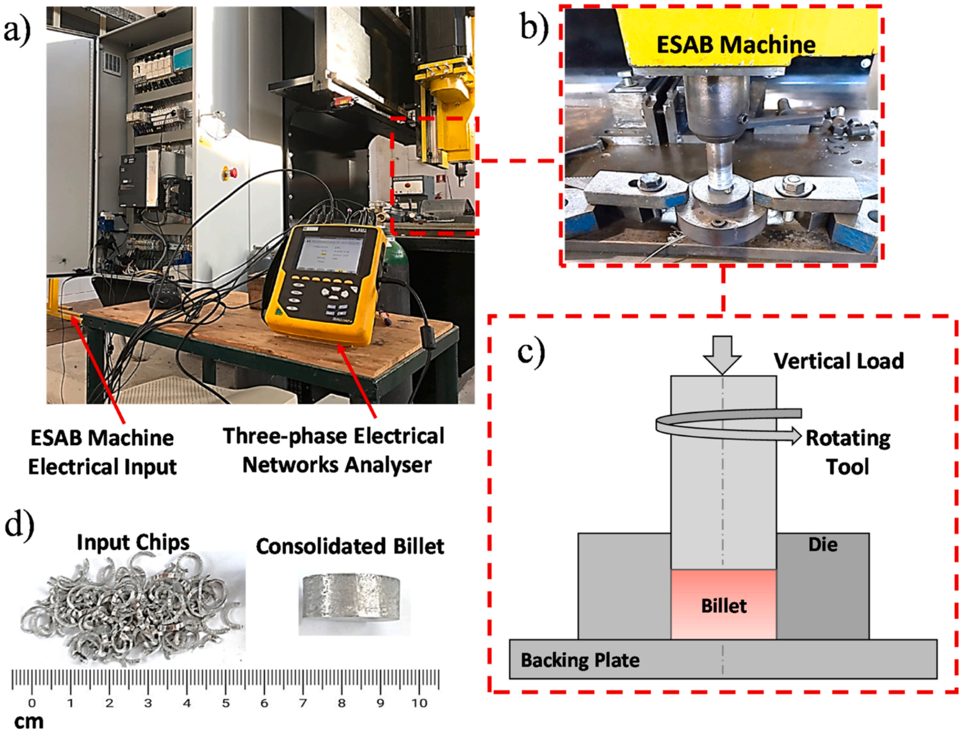

Fig. 1. (a) Electric power acquisition set-up; (b) enlargement of the FSC tooling, c) Sketch of the process, d) chips and consolidated billet.

is neither energy nor resource efficient. In this respect the main drawback for the conventional recycling processes is the permanent material losses occurring during remelting because of the oxidation [11]. In order to overcome such issue, researches have turned to Solid State Recycling (SSR) strategies. These approaches directly turn aluminum scraps into semi-finished products by avoiding the remelting step. The solid-state nature of such process category results in no material losses due to oxidation. Solid-state activation depends on pressure, temperature and contact time among surfaces to be joined. Thus, various solid state recycling techniques have been proposed based on the physical disruption and dispersion of the oxide contaminants by imposing significant plastic and shear strain. As a matter of facts, plastic deformation should be large enough to break the surface oxide layer of the metal in order to expose clean, non-oxidized metal surfaces and allow the formation of metallic bonds. The theory and mathematical modelling of solid bonding occurrence in case of aluminum alloys was presented by Copper and Allwood [12]. Wan et. al. [13] provide a comprehensive overview of the various SSR techniques for aluminum alloys recycling, and the authors cluster all the existing methods into two main categories: SSR techniques based on severe plastic deformation and SSR techniques based on powder metallurgy.

Concerning the approaches based on plastic deformation, various SSR extrusion-based processes have been investigated. In direct hot extrusion [14–16] the high extrusion ratio results in an increased level of introduced plastic strain, achieving bulk scrap-based profiles with comparable mechanical properties as the cast-based profiles. A variant of this approach was devised by Haase et al. [17]. In this paper the authors integrate extrusion with Equal Channel Angular Pressing (ECAP) for achieving superior mechanical properties in the recycled samples. Other authors applied screw extrusion [18,19] for recycling chips, in this approach the rotational movement generates extrusion pressure and introduces large shear strains that enhance the consolidation process. In this respect some authors have recently proposed the Shear Assisted Processing and Extrusion (ShAPE) do directly turn chips into hollow extruded profiles [20].

The Friction Stir Extrusion, is another extrusion based process used for recycling chips into wire/rod and was first presented by Tang and Reynold in 2010 [21]. In this process the friction between a rotating tool and the compacted chips to be recycled is used to produce heat and plastic deformation into the chips batch enabling the consolidation through backward extrusion. It is worth highlighting that extrusion based SSR processes have been already successfully applied to recycle Titanium alloys [22] and magnesium alloys chips [23].

Schulze et al. [24] suggested a combination of extrusion and rolling to obtain flat sheets out chips ready to be formed by Deep Drawing processes. Cooper et al. [25] proposed chips extrusion as a preparatory step to remelting in order to save energy and resources during aluminum chips recycling.

As far as the powder metallurgy-like approach is concerned, spark plasma sintering has been successfully applied to recycle both aluminum [26] and magnesium alloys [27] chips.

Other authors presented hybrid approaches: Kohck et al. [28] proposes a recycling process made of a field-assisted sintering (FAST) process to consolidate the aluminum alloys chips, and a forward rod extrusion process. Behrens et al. [29] recommended a hybrid approach made of compacting, sintering and forming to consolidate aluminum chips.

Friction Stir Consolidation (FSC) is one of the SSR processes that has effectively recycled machining chips into solid blocks/billets [30]. Actually, a rotating tool is pressed onto chips which are confined in a billet chamber. The heat generated by friction and plastic deformation results in softening of the material. Undergoing rotation and compression, the interparticle gaps are diminished and then the softened particles are welded together and consolidated. The authors proved that such processes are more energy efficient with respect to conventional remelting based recycling route [31].

Although fully consolidate billets can be obtained, these are characterized by non-homogenous mechanical properties and microstructure [30–33]. Recently, some of the authors of the present paper have successfully introduced new strategies, based on multi-step approaches, aimed at obtaining billets with more uniform mechanical and microstructural properties [32].

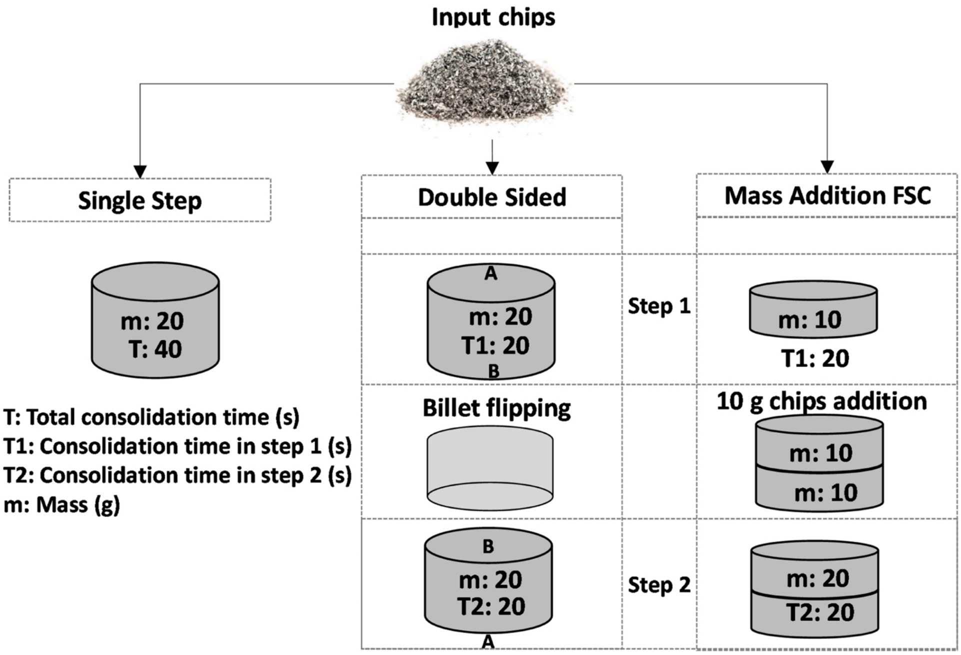

Fig. 2. Sketch of the three FSC variants.

Although the authors developed a preliminary primary energy demand comparative analyses [31], a full environmental impact analysis on single step FSC by means of LCA has not been developed yet. Also, the multi-step variants, recently proposed by the authors [33], worsen the environmental impact of FSC processes as they require a higher amount of electric energy, in consequence the environmentally friendly nature of such an approach is still to be proved.

The present paper aims at covering both of the aforementioned research gaps. Specifically, a full LCA comparative analysis is presented: the environmental performance of single step, multi-step FSC processes and remelting based routes are quantified and compared to one another. Also, different scenarios are analyzed to provide guidelines for the correct implementation of FSC based recycling processes.

The structure of the paper follows the ISO 14040 structure; as a matter of fact, after presenting the FSC process in the materials and methods section, the results are all reported in the section named LCA framework.

2. Materials and methods

The starting material was AA2024-O round bar with 30 mm diameter and its composition by mass percentage was 94.1Al-0.003-Cr-4.26Cu-0.57Fe-0.004Mg-0.01Mn-0.129Si-0.003Ti-0.008Zn-0.5Pb-0.12Sn. The bar was reduced into chips by COMEV 180 turning machine with a rotational speed of 460 rpm, feed rate of 2 mm/revolution, depth of cut equal to 1.5 mm and 15° cutting angle (the obtained chips are reported in Fig. 1d). The chips were properly cleaned by acetone, which is commonly used as a washing solvent for cleaning metal scraps and is equally applicable both at laboratory and industrial scales. The chips were loaded in H13 cylindrical steel die with a nominal diameter of 25 mm and compacted by 5 kN force through a H13 cylindrical steel tool with a 25 mm diameter. The die fixture and tool were coupled with ESAB LEGIO 3ST, a dedicated friction stir welding machine (Fig. 1).

To be specific, the experimental setup of previous research [33] was considered.

The case study was focused on two variants of friction stir consolidation: single step and multi-step approaches with process parameters 2000 rpm tool rotational speed, 40 s process time, and 20 kN consolidated force. A sketch of the FSC concept and related multi-step variants are reported in Fig. 2. In the multi-step approach, two sub-types were applied: mass addition and double-sided methods.

These methods of FSC and the aforementioned process parameters were selected considering both the efficiency in electrical energy consumption during the FSC process as well as the mechanical performance of the obtained billets. Multi-steps variants allow the hardness and grain size distribution homogeneity to be improved with respect single step approach [33].

2.1. Process

A billet was manufactured from a 20 g input chips batch. During single step FSC, the total input mass (20 g) was loaded inside the die and compressed with 5 KN pre-load. In the subsequent step, tool rotation was activated, and the pressing load was gradually increased from 5 kN to 20 kN (consolidation force) in 30 s and this step was referred to as transition phase. Due to machine limitation, it was not possible to instantly increase the load from 5 kN compaction force to 20 kN consolidation force, but rather it was gradually increased with a step increment of 0.5 kN/s. Upon reaching the desired load, finally, the whole charge was consolidated at 20 kN constant force for the processing time of 40 s. In short, the whole process was completed in two steps: the transition phase, and the consolidation phase.

On the other hand, the multi-step approach has almost all parameters in common with single step FSC, such as 2000 rpm tool rotating speed and 20 KN consolidation force. But the only exception was the number of steps of the process executions (see again Fig. 2). Specifically, during the mass addition method, half of the input mass (10 g) was consolidated for half of the total process time (20 s), and then the remaining chips mass was added in the next step and consolidated for the remaining half of the processing time. On the other hand, in the double-sided method, the whole chips batch was consolidated for half of the processing time in the first step, then the billet was removed and reentered in the die upside down. In the final step, the consolidation step was applied for the flipped side of the billet for the remaining half of the processing time. In Fig. 2a sketch of the analyzed FSC variants is reported.

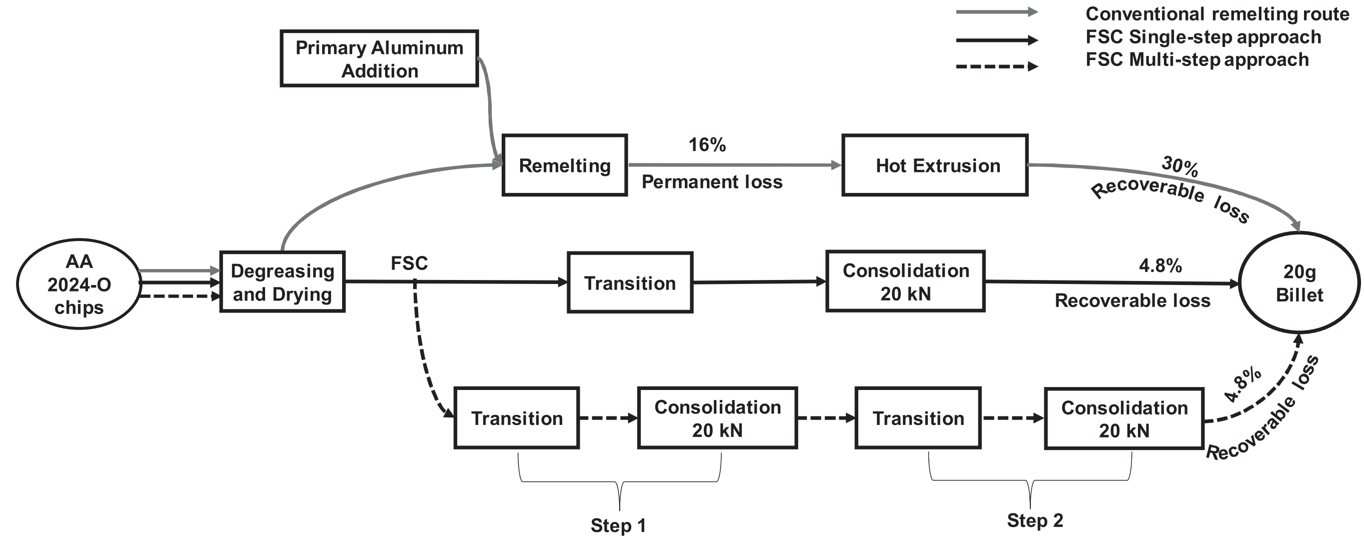

Fig. 3. The adopted system boundary.

2.2. Analyzed output

The hardness distribution and the microstructure of the obtained samples were analyzed in a previous paper [13]; the power-time data was collected by connecting a three-phase electrical networks analyzer to the input electrical connection of the ESAB machine (Fig. 1a). The power was measured for the productive step (consolidation) as well as for transition phase (Fig. 4). Energy consumption was then calculated from the power-time graph by direct numerical integration method.

3. LCA framework

The LCA methodology followed in this study is described in this section, which includes the definition of the goal and scope, functional unit, systems boundaries, the life cycle inventory analysis for the different recycling routes. The interpretation of the LCA results is presented in Section 3.4.

3.1. Goal and scope definition

The aim of the LCA is to characterize the environmental performance of an innovative solid state based aluminum alloys recycling process. The LCA results can be used to compare the environmental impacts of conventional remelting based route with two different variants of the FSC based process. The aim is to analyze these performances in different production scenarios. As a matter of fact, the LCA comparative analyses were developed with varying both the electric energy demand of FSC processes and the energy mix in the electricity supply. As far as the electric energy demand is concerned, two different scenarios were considered: 1/including the entire electrical energy absorbed during the transition phase (worst scenario); 2/ leaving the contribution of the transition phase out of the analysis (ideal scenario).

This choice was driven by the awareness that the used experimental procedure, including the transition phase, is to be considered a lab scale procedure and it is not optimized for industrial practices. On the other hand, not considering the transition electrical energy at all, is for sure an

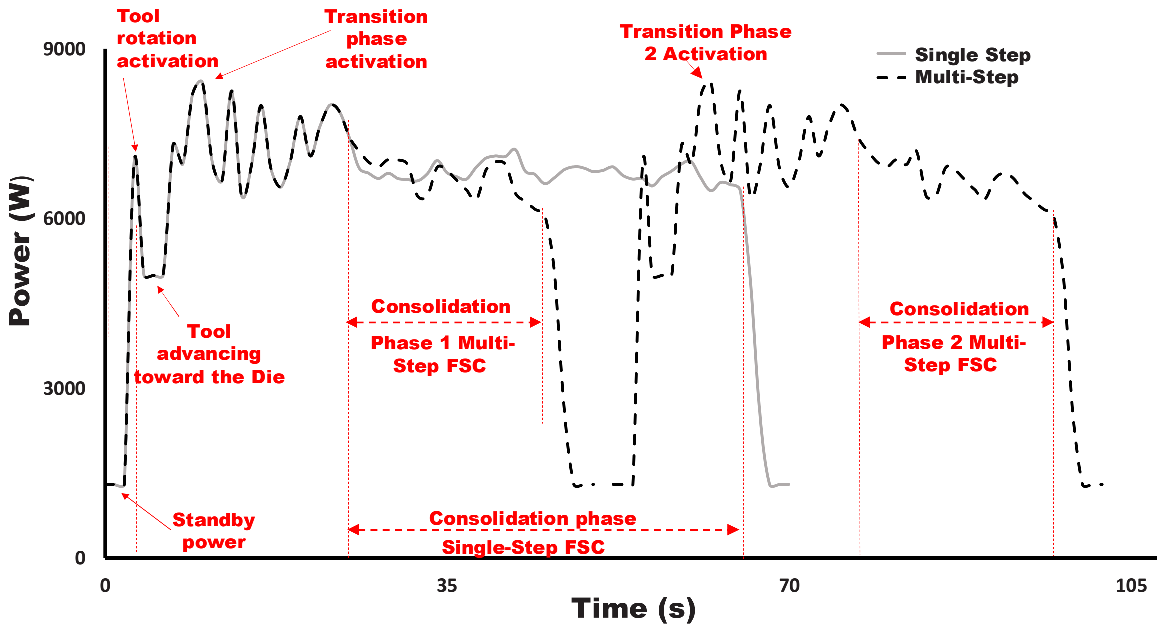

Fig. 4. Electric power trend of single and multi-step FSC.

Table 1
Electric energy demands of FSC processes.

	Transition energy (MJ)	Consolidation energy (MJ)	Total energy (MJ)	Specific energy (MJ/kg)
Single Step	0.178	0.272	0.450	22.50
Multi-Step	0.355	0.267	0.623	31.16

unrealistic, ideal, scenario. Analyzing both of them allows to have a clearer picture of the impact of such phase with respect the overall comparative LCA analysis. Thus, the LCA study can provide guidelines for the sustainable recycling processes selections in different scenarios.

3.2. Functional unit

The functional unit selected to develop the analysis is the production of 20 g of AA2024-O aluminum alloy billet starting from chips.

Specifically, according to the die geometry reported in Section 2, the considered billet is a cylindrical one with a diameter equal to 25 mm and a height of 15 mm. It is worth remarking that the size of the billet is the maximum obtainable with the installed power and loads of the used machine. In fact, as proved by the authors, larger amount of chips would require higher vertical load to be applied, which is not available in the used ESAB machine [33].

3.3. System boundary and Life Cycle Inventory (LCI)

As the aim of the LCA study was the characterization of the environmental performance of single and multi-step Friction Stir Consolidation, a full comparative analysis with the conventional (remelting based) route is proposed. All the process steps for turning chips into billets are considered for each route as reported in Fig. 3.

Degreasing and drying have been envisaged for all the routes and the processes have been modeled by using data from Duflou et al. [11] and Cooper et al. [25].

Concerning the remelting route, besides the melting step which includes also casting and alloying, hot extrusion and sawing were considered to obtain the desired billet. Both the remelting and the hot extrusion were modeled by using the Life Cycle Inventory developed by the EEA [35].

As mentioned in introduction, permanent material losses occur while remelting due to oxidation. In this research it is assumed that the amount of material losses is as high as 16 %; this amount is the most likely scenario to happen during chips melting as suggested by Duflou et al. [11]. The impact of permanent material losses was considered by adding the same amount of primary aluminum in the model. The primary production of AA2024-O was modelled in SimaPro.

All the recoverable losses reported in Fig. 3 have been modeled as usable material input for the process the scraps have been generated from. In other words, it is assumed that FSC process scraps are used together with unprocessed chips as input material for the billet production; the same approach was adopted for modelling the hot extrusion process.

As far as the electrical energy demand of Friction Stir Consolidation is concerned, it was experimentally measured as described in the materials and methods section. In Fig. 4 the analyzed power trends of both single step and multi-step approaches are reported. All the different production modes of the FSC single step and multi-step working cycles are reported.

No relevant difference in terms of power trends were observed between mass addition and double-sided strategy, instead. Therefore, from now on, “Multi-Step FSC” expression will be used and this will refer to both Double sided and Mass addition approaches.

It is worth remarking that, likewise, in the single step approach, each

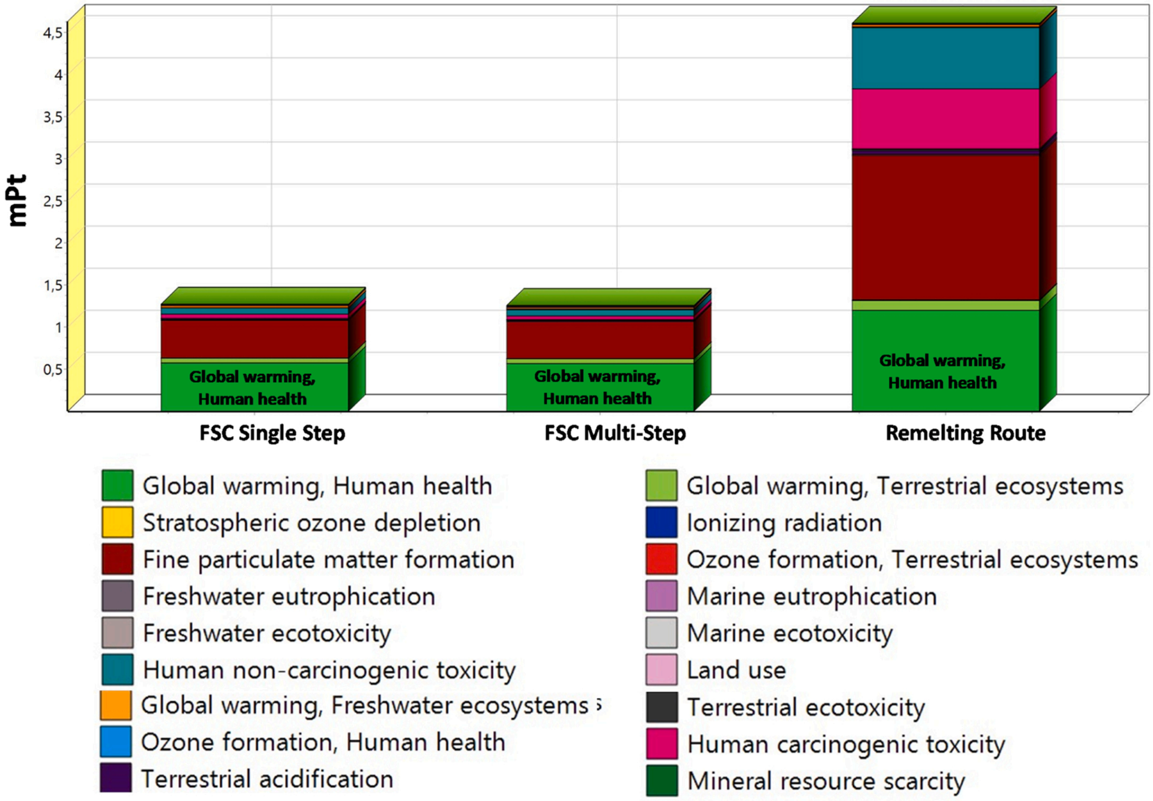

Fig. 5. LCA results (mPt) for the Ideal Scenario (Italy).

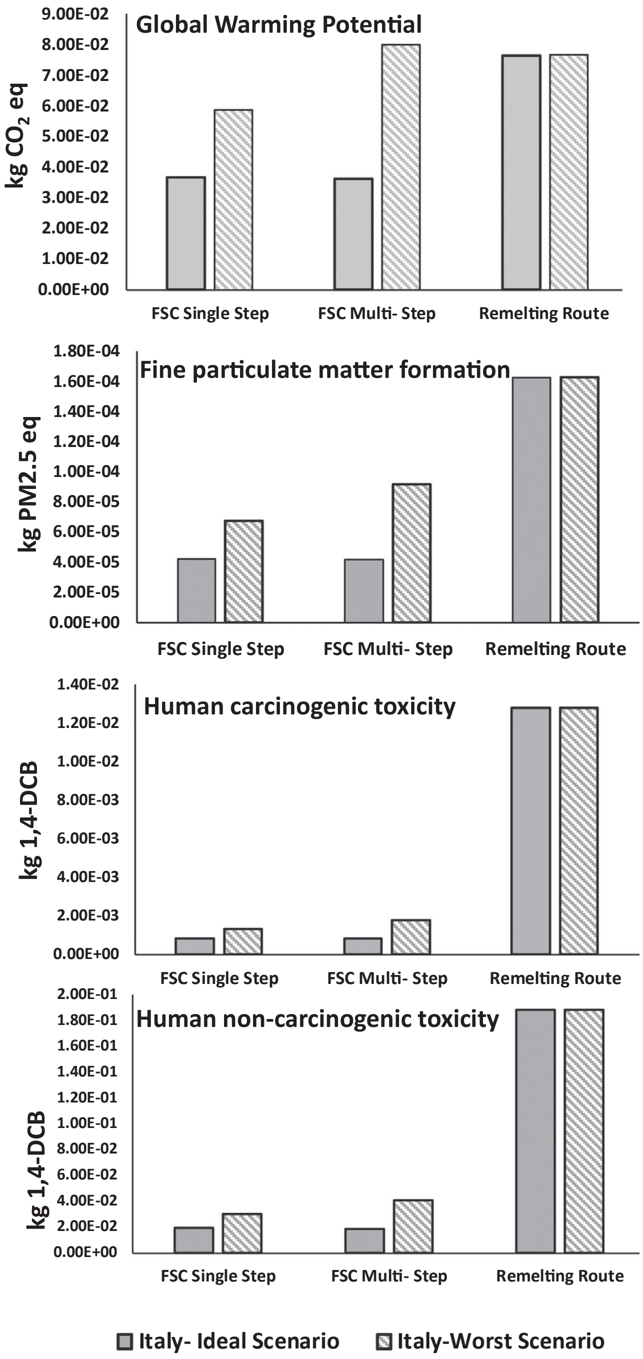

Fig. 6. LCA results (midpoint) for Ideal and Worst scenarios (Italy).

step of the multi-step procedure has two phases: transition and consolidation. However, the productive step's processing time is equally divided between the steps (i.e., 20 s consolidations time during each step). Thus, multi-step FSC is comprised of two transition and two consolidation phases (see Fig. 2 and Fig. 4). Overall, during the productive/consolidation production mode a slight decreasing trend can be observed, this is due to the increase of temperature while consolidating the chips. After recording the power trend over multiple experiments, the energy was calculated as described in Section 2.2, the main results of energy demand are summarized in Table 1.

3.4. *Interpretation of the results*

In order to identify the impact categories to comment on for describing the environmental impact of each recycling processes, the Endpoint ReCiPe2016 (H) analysis was developed. In Fig. 5 the analysis, selecting the Italy energy mix in the electricity supply and the ideal scenario (with transition phase not included in the analysis), are reported.

In Fig. 5 the contribution of each impact category towards the total mPt (milli-points) is depicted. It is possible to notice that the remelting step has the highest environmental impact while the FSC variants are characterized by a low and very similar environmental impact. Fig. 5 reveals that the main impact category contributors are the Global Warming Potential (GWP), the fine particulate matter formation, the human carcinogenic toxicity and human non-carcinogenic toxicity. Since the single score Endpoint has no physical meaning and the overall value is affected by the normalization or weighting procedure, the four aforementioned categories will be selected in the next sections for discussing the results with varying the analysed scenario.

3.5. *The impact of the transition phase of the FSC processes*

In this section the results of the four identified impact categories have been analyzed.

Specifically, for the FSC single step and multi-step the results for both the ideal scenario (transition phase not included) and the worst scenario (the whole transition phase included in the analysis) are reported. Italy is selected as country for the energy mix in the electricity supply. In the graphs of Fig. 6, for the sake of clarity of representation, two different bars are reported also for the remelting route although the values for the two scenario are exactly the same. For the ideal scenario there is no relevant difference between the single and the multi-step FSC approach. Also, the FSC process is by far the best solution for all the four analyzed categories. Whereas, when the worst scenario is analyzed, visible differences can be seen between the two variants of the FSC process. Actually, the performance of FSC process worsens for both of the variants in all the categories. Moreover, the performance of multi-step approach is significantly worse than that of the single step in all the categories. What is more, for the Global Warming Potential the worsening is so substantial that the multi-step approach is no longer better than the remelting route. It is worth remarking that the worst scenario is an unrealistic one and, despite that, the GWP values are very close each other; therefore, FSC is an environmentally friendly solution also in very pessimistic circumstances.

3.6. *Analysis with varying the energy mix in the electricity supply*

In order to analyze the impact of the energy mix in electricity production on the environmental performance of FSC recycling process, the LCA was performed considering the energy mix of three different countries: Norway, Italy and China. Norway was selected as it is a country where renewable sources represent a significant share of the electrical energy production sources. China is selected as an example of a country that heavily relies on fossil fuel burning with a significant share allocable to coal. Italy could represent an average scenario as the majority of the countries are in the middle of the transition process towards a massive use of non- renewable sources. The results for the four analyzed categories are reported in Fig. 7.

As it is possible to see, the China scenario provides the worst performance for any category and process selected. Also, as FSC relies basically on electrical energy consumption, for the GWP indicator both single and multi-step versions provide worse values than the remelting route, the same trend can be noticed for the fine particulate matter formation category. For the two other analyzed categories both of the FSC variants outperform the remelting route, instead. When the Norwegian scenario is considered, the FSC environmental impact improves significantly becoming by far the most environmentally friendly option. In order to further explore the impact of the country on the comparative analysis results, Cumulative Energy Demand (CED) analyses for China

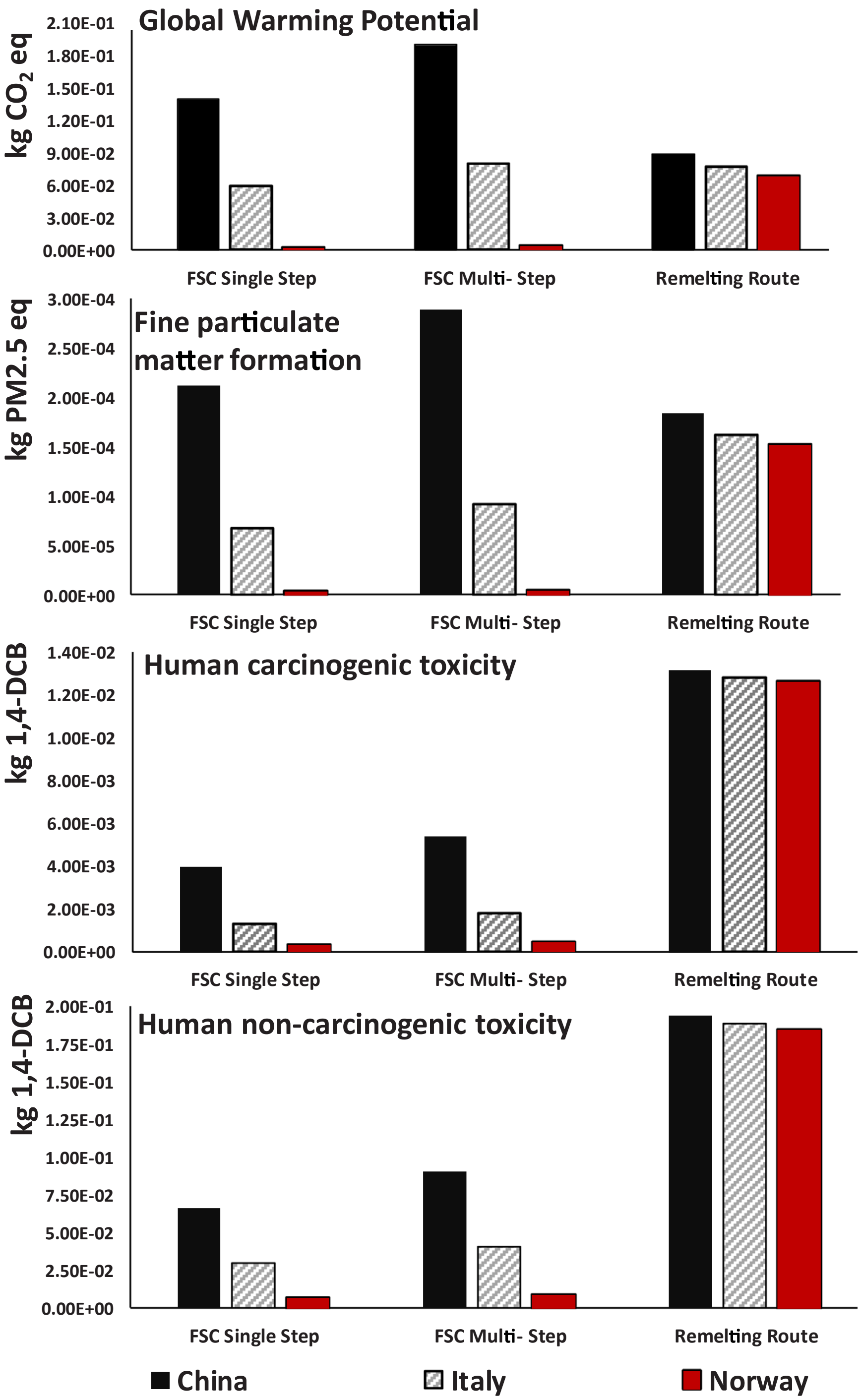

Fig. 7. LCA worst scenario results (midpoints) for different countries.

and Norwegian case studies (worst scenario) were developed. Results are reported in Figs. 8 and 9, respectively. It is possible to see that the CED values decrease significantly when Norway scenario is selected. It is worth highlighting that, for the sake of clarity, in Figs. 8 and 9 MJ was selected as CED unit for the China scenario whereas KJ was used for the Norwegian one. Moreover, the contribution of each energy source towards the total is visible in the graph, the strong dependency on fossil fuel combustion of China and the large share of renewable sources for the Norwegian case study are clearly visible in Figs. 8 and 9, respectively.

4. Discussion of the results

The developed analyses revealed that FSC, especially in its single step version, allows substantial environmental impact reduction with respect conventional recycling route. However, the environmental performances are affected by the non-productive transition step as well as by the energy mix in electrical energy production. As a matter of fact, the multi-step version turned out to be the worst approach when the transition step is included and China scenario is analyzed. Although the worst scenario is an unrealistic one, it deserves to spend some words on it. In fact, it is worth highlighting that for a green implementation of FSC aluminum alloys recycling processes, it is needed to develop dedicated

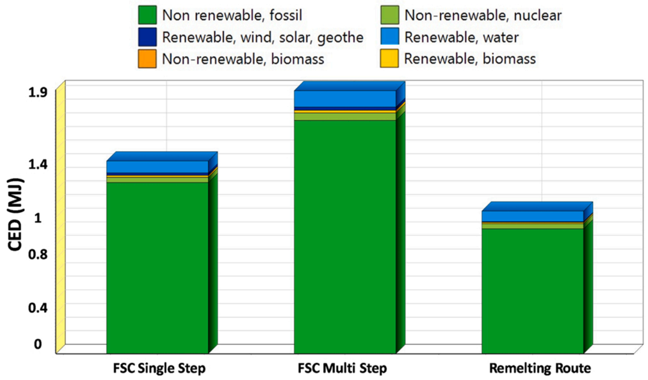

Fig. 8. CED worst scenario results (China).

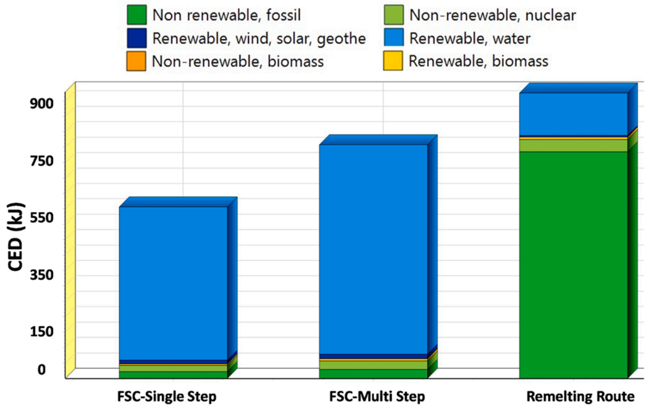

Fig. 9. CED worst scenario results (Norway).

Table 2
Properties comparison of different recycling routes.

Properties	Single step FSC	Multi-step FSC	Remelting route
Risk of uncompleted consolidation	High (at the bottom-outer region)	Extremely low	N.A
Hardness and grain size distributions Homogeneity	Poor	Very good	Excellent
Environmental Impact	Very low	Depends on the analyzed scenario	High
Process complexity	Very Simple	Simple	Extremely complex

machines equipped with technological solution able to minimize the transition step.

Also, as proved by the authors in previous analyses [33], multi-step approaches allow a better billet, in terms of hardness and grain size distribution, to be obtained. Therefore, in a multi-objective perspective a higher environmental cost might be compensated by a superior quality of the recycled component.

In order to better discuss the different properties characterizing the three recycling approaches here analyzed, the Table 2 is reported. In this table a qualitative description of quality, mechanical and environmental properties is reported.

It is worth remarking that the single step FSC is characterized by an axisymmetric heat flow moving from the top toward the bottom. As already proved by Tang and Reynolds [34], the die and the backing plate serve as a heat sink, reducing the temperature and, more importantly, constraining the deformation/flow of nearby aluminum alloy chips. As a consequence, the further the chips are from the top (rotating tool) the more difficult are the solid bonding conditions to be achieved. The greatest constraint on deformation is obtained in the bottom/down region (intersection of the billet die wall and the back plate), where lack of consolidation is often observed. Lack of consolidation is particularly significant when too high billets (with respect the available vertical loads of the used machine) need to be manufactured.

This is the reason why the multi-step approach is preferable when the height of the required billet cannot be produced with single step or when homogenous properties obtainment is a strict production constraint to be met.

5. Conclusions

In this research the environmental impact on an innovative recycling route for aluminum alloys chips is presented. The paper, by integrating unit process level analysis and Life Cycle Assessment of the entire recycling route, allows the environmental performance of Friction Stir Consolidation recycling approach to be characterized.

Results revealed that FSC allows substantial environmental impact reduction with respect to conventional (remelting-based) recycling route. On the other hand, the research pointed out the urgency to still optimize the process to increase the Technology Readiness Level (TRL) of the proposed approach; specifically, two main improvement directions can be identified: 1/ the necessity to improve the mechanical properties provided by the single step version of the process, 2/ the need of reducing the electrical energy demand of the non-productive mode (transition phase).

Concerning the first improvement direction, the implementation of an energy efficient heating system will be studied in the future by the authors. The main idea is to heat up the chips batch in the die in order to improve the consolidation at the bottom of the billet already in the single step version.

As far as the reduction of the non-productive energy demand is concerned, technological innovation at machine tool architecture is to be studied in order to reduce the time and the energy of the transition step. More in general, it would be necessary to design a dedicated machine for FSC based recycling processes, this would further reduce its environmental impact.

Also, the suitability of the obtained billet to be used for forging and machining operations will be tested, this would allow to further characterize the industrial applicability of the proposed approach.

Finally, as discussed in the discussion of the results section, the analyzed recycling processes variants are characterized by conflicting performance (see again Table 2); further development will concern the implementation of decision support tools based on Multi Criteria Decision Making (MCDM) methods able to take into account simultaneously mechanical, environmental, economic and productivity design objectives. This would allow the best recycling process to be identified with varying the production scenario.

One last important finding of the present research is that the environmental performance of FSC, with respect to the remelting route, strongly improves with increasing the share of renewable energy sources in the energy mix for the electric energy production. Therefore, in the upcoming future, when the share of the renewable sources will be wider, such kind of process would allow further and substantial environmental impact reduction in aluminum alloys recycling sector.

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