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Integrating an ageing model within Life Cycle Assessment to evaluate the environmental impacts of electric batteries

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The electrification of vehicles is seen nowadays as a promising way to decarbonize the personal transportation. The assessment of environmental impacts of electric batteries is usually case-specific due to the complex modelling of such systems which present a large variability in designs, in user behavior or in geographic use conditions. A typical example is the battery lifespan which is arbitrarily chosen in most cases, even though it has a decisive influence on lifecycle emissions. Computing the battery lifespan in addition to a Life Cycle Assessment (LCA) would enable to highlight new hotspots and new parameters to reduce the environmental impacts of batteries. This paper introduces a new approach, based on a LCA conducted with the open-source software Brightway and built on primary data collected from a complete disassembly of a commercial electric vehicle. An original functional unit has been proposed that better represents the service offered by the battery over its lifetime and a semi-empirical ageing model has been integrated to predict more precisely the battery lifespan depending on design parameters and the use conditions. This innovative methodology is easily parameterized and aims to compare several eco-design strategies.

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Keywords: Lithium-ion battery; Life Cycle Assessment; Eco-design; Climate Change; Ageing model; Environmental impacts; Optimization**Introduction**

In 2020, the market of lithium-ion batteries (LIB) reached 230 GWh of capacity. The automotive market was the largest application (69%) and its share has considerably increased this last decade since it was less than 1% of LIB market in 2000 [1]. Reduce the environmental impacts of an electric vehicle (EV) requires the eco-design of electric batteries since it accounts up to 41 % of the total greenhouse emission gas (GES) of a EV production [2]. Plenty of Life Cycle Assessments (LCA) have been led on electric batteries these last years [3]–[10]. Most of them focus on the production phase [11] and on the climate change impact category [4], [11], [12]. Results range from 53 kg CO₂ eq/kWh to 313 kg CO₂ eq/kWh [4], [11]–[13]. As highlighted by several authors, a large variability appears between the studies due to the use of several functional units,

boundaries, impact categories, but also due to the lack of primary data [14]–[17]. This comes from the nature of the battery itself, which appears to be a “complex system” which is characterized by a large number of interdependent sub-systems, of numerous alternative solutions, with several operating modes, and which evolves during its lifetime by interacting with its environment [18]–[20]. Therefore, assessing the environmental impacts of LIBs is usually case-specific due to the complex modeling of such systems made of a large variability in designs, in user behaviors or in geographic use conditions.

Another issue of LCAs of electric batteries lies in the high sensitivity of the results. One of the major sensitive parameter is the lifespan of the battery, as noticed by several authors [6], [7], [11], [16], [21]–[23]. Indeed, the lifespan is arbitrarily chosen in most studies: usually 10 years or 150000 km.

However, in a cradle-to-grave approach, this value is used for normalization and can thus, greatly influences the final results [22], [24]. Arshad et al. [11] show that emissions from the same study can be extended from 9.86 g CO₂ eq/km to 72–80 g CO₂ eq/km when the lifespan is decreased from 300000 km to 150000 km.

Several studies attempted to bring agility to their methodology. Cox et al. [25], [26] develop a parameterized model for the whole electric vehicle and build scenarios to assess the benefits of autonomous vehicles, and the influence of the future electric mix. This work has recently been reused by Sacchi et al. [27] to build an interactive tool *calculator*, which shares the same systemic approach than the Excel spreadsheet from the Argonne National Laboratory *Everbatt* [27], [28]. Egede [20] takes also advantage of a parameterized Life Cycle Inventory (LCI) to study the influence of the use conditions, the geographical conditions, and the lightening strategies. However, none of these studies incorporates the lifespan issue. With the ‘Dualfoil’ model, Lybbert et al. [17] consider simple ageing model to assess the influence of cell properties (thickness, porosity, ambient temperature, discharge rate) on the environmental impacts. Ma et Deng [29] go further by optimizing the cathode thickness and the bilayer number over the whole lifecycle of the battery thanks to a kinetic model. Nevertheless, these studies only focus on the cells and do not consider the battery pack as a whole system.

This paper offers a more accurate method to assess the environmental impacts of electric batteries by integrating an ageing model to LCA in order to predict the lifespan depending on the design parameters. This model is easily parameterized and aims to be used as an eco-design tool for the battery pack. The details of the ageing model are presented in a first part, followed by a section that explains its integration into the LCA approach. The final part provides the first results that have been obtained through this methodology.

1. Ageing model

Previous internal projects have measured capacity losses for several commercial cells under different operating conditions. This work ended up with the construction of an ageing cartography for a LiNi_{1/3}Mn_{1/3}Co_{1/3}O₂ cell (NMC111). Calendar losses, related to the period where the battery is unused, and cycling losses, that occurs during the battery charge and discharge, are measured at different ‘breakpoints’ which refers to the following parameters of the cell: the temperature, the State of Charge (SoC), the current (or the C-rate), and the previous loss capacity. Cells are tested at 5 different temperatures, namely -10 °C, 10 °C, 25 °C, 45 °C and 60 °C, with 7 different SoC window, namely 0-100 %, 0-80 %, 0-90 %, 10-100 %, 10-90, 20-100 %, 20-90 % at 5 different charge/discharge rates (commonly called C-rate), namely 0.30/3C, 1C/1C, 2C/2C, 3C/3C, 4C/4C. A 1C discharge means that a fully charged 1Ah battery should provide 1Ah for one hour. A 2C discharge would last only 30 minutes.

This results in a cartography that contains all the measured capacity losses with the associated breakpoints. The Fig. 1 illustrates what can provide the cartography for the cycling losses depending on only 2 parameters: the cell temperature and the SoC. This cartography is then interpolated so that, the

total capacity loss – the sum of the calendar and the cycling losses – can be computed at any breakpoints. A similar approach has been proposed by Montaru et al. [30] to build a calendar ageing model.

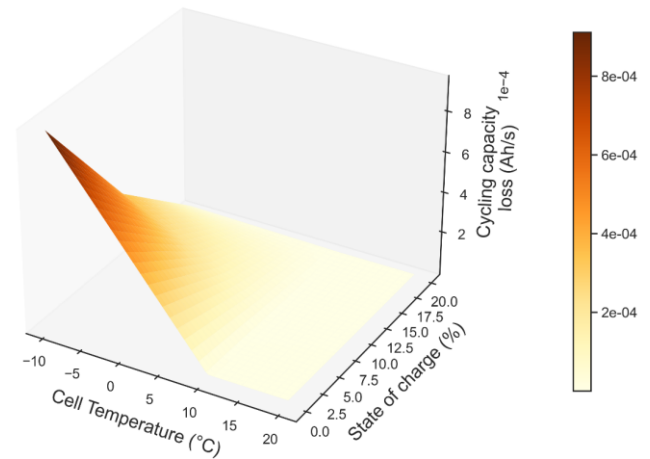


Fig. 1. 3D graph extract from the cartography: variation of the cycling capacity loss depending on the cell temperature and the State of charge (during a 1C discharge)

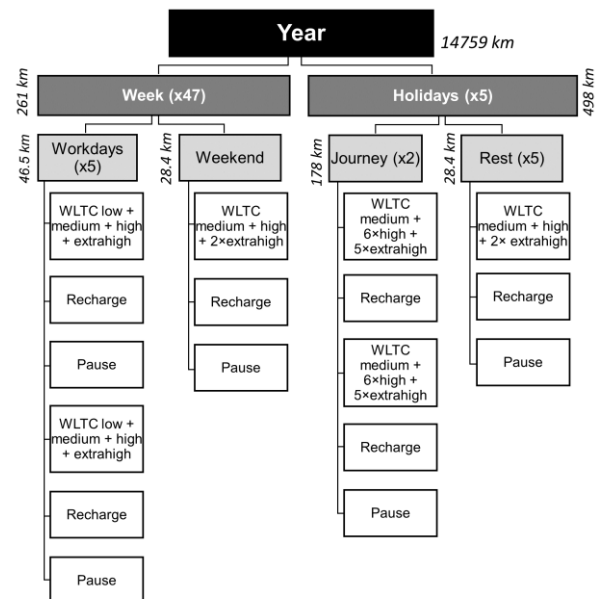


Fig. 2. Use profile for the use of an small-size EV with a 33.3 kWh battery pack during one year

The use profile described in Fig. 2 is used in parallel with the cartography to compute the capacity loss at any step of the profile. It is constructed over a whole year to represent an annual intense use of a small-size EV. It is based on the standard Worldwide Harmonised Light Vehicles Test Procedure (WLTP) which relies on standardized driving cycles (WLTC). These cycles are subdivided into 4 phases which simulate an urban, semi-urban, rural and highway driving scenario. During the workweek, a complete WLTC, which is equivalent to 1800 seconds and 23.266 driven kilometers, is performed to go to work and go back home. During the

weekend, the car is used to drive 28.4 kilometres on highways, rural and semi-urban sections. Five holiday weeks are considered (the 1st week of April, the two first weeks of August, and the last two weeks of December). To simulate an intense holiday journey, 178 kilometres are driven the first and the last day of the holiday week (referred as ‘Journey’ in Fig. 2) mainly on highways. The same profile as the one from the weekend is used for the other days of the holiday week. In a year, 14759 km are driven through this profile, which is a bit higher than the average of French drivers (12200 annual km) [31]. Thus, this use profile can be considered as a severe intensity use.

To get the cell temperature T_{cell} , a simple electrothermal model is used to represent the internal heating by Joule effect and the thermal exchanges with ambient temperature:

$$mc \frac{dT_{cell}}{dt} = R_{i,cell} \cdot I_{cell}^2 + hS(T_{ext} - T_{cell}) \quad (1)$$

m is the cell mass (kg), c the specific cell heat capacity ($J.K^{-1}.kg^{-1}$), h the heat transfer coefficient ($W.m^{-2}.K^{-1}$), S the external cell surface (m^2), $R_{i,cell}$ the internal cell resistance (Ω) and I_{cell} the cell current (A). This representation assumes that the battery pack has a homogeneous temperature distribution and that the cells exchange thermally directly with the ambient exterior on all the cell surface. The equation (1) is numerically solved through an implicit resolution to get T_{cell} . The numeric values of the parameters cannot be detailed for confidential reasons. A more complex model will be integrated in later works to better represent the temperature behaviour of a battery pack. Once we have access to the cell current, the cell temperature, and the SoC at each step of the use profile, it is now possible to compute the capacity loss at any time of the battery lifetime. Thus, we are able to compute a time-dependent function from this capacity loss $E_{aged} = f(t)$ that enables to predict the battery lifespan.

2. Integrating the ageing model into LCA

The scope of the study is the battery system which includes the casing, the modules, the cooling circuit, the powerbox and the power electronics. On the contrary to most studies, the LCI of the battery takes into account the power electronics, which contains the inverter, the DCDC controller, and the charger. Since these elements interact directly with the battery and have an influence on ageing, we include these elements in our scope. The boundary of the LCA is cradle-to-grave, which goes from the extraction of the raw materials, the production of the battery, its use during its lifetime and its End-of-Life (EoL). As the scope of the study is only the battery, the consumption of the car without the battery nor charger, is subtracted to the total energy required to recharge the battery.

The functional unit (FU) aims at describing the service offered by the object. As recommended by the IPCC Handbook, it shall answer the questions ‘what’, ‘how much’, ‘how well’, and ‘for how long’ [32]. Thus, concerning an automotive electric battery, the FU can be stated as follows : to provide the energy to the car to perform the use profile during one year. Every year, we check if the energy stored in the battery which is computed from the ageing model, is sufficient to perform the use profile during one year, that is to say, if the state of charge reaches 0% during the year. It is usually admitted that the battery has come to its EoL when 70 to 80% of the initial

energy remains. However, Saxena et al showed this threshold has nothing to do with the real use of the drivers, since more than 85% of the american daily trips can still be performed at 80% SoC [33]. Linking the EoL to the use profile makes possible to know exactly when the battery cannot fulfill the user needs instead of arbitrary choosing one threshold or one reference lifespan.

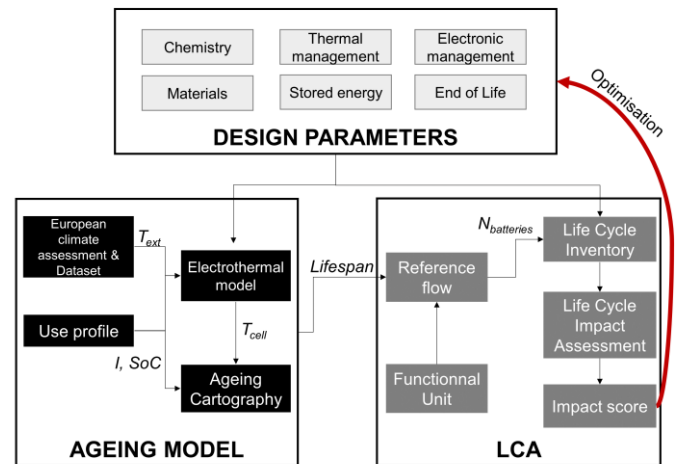


Fig. 3. Structure of the model that integrates an ageing model within LCA and which enables to assess the impact score of several battery design

The reference flow to meet the FU refers to the number of battery that is necessary to perform the use profile during one year. This value is assessed by dividing one year with the predicted lifespan, computed thanks to the ageing model. We assume this number can be decimal: the main purpose is to establish a reference baseline that allows the comparison of the impact score of several design configurations. A higher lifespan will lead to a smaller reference flow, which will reduce the impact score.

The main asset of this method lies in its ability to take into account the influence of the design parameters not only on the Life Cycle Inventory (LCI) but also on the lifespan since it will change the breakpoints of the ageing model. Therefore, from several design configurations, it is now possible to compute a lifespan and a reference flow for each set of designs, that leads to different impact scores as shown in Fig. 3. The lifespan is not seen as an independent input parameter anymore but is directly estimated from the design and the use conditions.

3. Results

3.1. Cradle-to-gate score

The LCA is based on an EV with a 33.3 kWh battery pack that weighs 283 kg, and contains 96 prismatic cells $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$ (NMC111). The cells have a 94 Ah capacity and are supposed to be manufactured in China. A Bill of Materials (BoM) is obtained from a whole disassembly of a small-size EV that gives the amount and the composition of each subcomponents of the battery pack. The EoL is modelled as a hydrometallurgy process with mechanical pretreatment (LithoRec process). Avoided impacts from the recovery of

energy (during battery discharging), and the production of recycled cobalt sulfate, nickel sulfate, manganese sulfate and lithium carbonate are taken into account. Metals such as aluminium, steel, copper but also plastics and electronics are supposed to be wastes. The LCI is completed by the database EcoInvent 3.8 for the background data. All computations are performed in Python 3.9 with the open-source LCA software Brightway2 [34]. The study will focus for now, on the impact category Global Warming Potential (GWP) of the methodology Environmental Footprint (EF) v3.

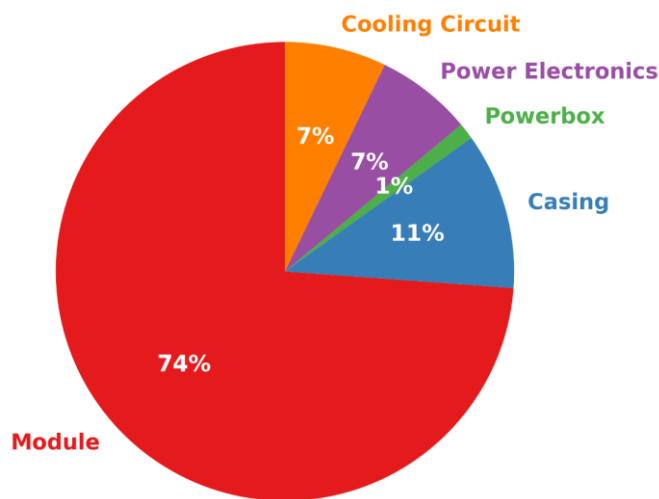


Fig. 4. Mass share of the studied battery system

As shown in the Fig. 4, active materials – cells in the modules – are the first contributor in the mass share with 74 % wt of the total mass followed by the casing (11 %). This is in line with the literature : Dai et al. [3] consider a battery with 73 % wt of cells higher than the 55% wt of cells for Kim et al. [9].

Fig. 5 represents the cradle-to-gate environmental impacts for the production of one kWh of battery in the impact category climate change. Contribution analysis is also presented in the Fig. 5 to highlight the relative share of each subcomponents. The LCA score of this study is 155 kg CO₂ eq/kWh and the modules are the major contributor in this impact category. This belongs to the upper range of literature results: 140 kg CO₂ eq/kWh was obtained by Kim et al. [9], a mean of 110 kg CO₂ eq/kWh by Peters et al. [4] or even lower with 73 kg CO₂ eq/kWh by Dai et al. [3]. This can be explained by the contribution of the power electronics, which is not integrated in the aforementioned LCAs, and which accounts in our study for 16 % of total Greenhouse Gas (GHG) emissions for the production phase. Besides, these studies are based on different data sources and assumptions that lead to an inherent variability of the results.

3.2. Capacity loss

Fig. 6 shows the result of the simulation of the loss capacity during 20 years. The mean consumption of the vehicle when driving is 0.1779 kWh/km. It can be noticed that the capacity drops highly in the first year and decreases in a linear way from 5 years. At 10 years, the useable energy stored in the battery

amounts to 71.2% of the initial energy.

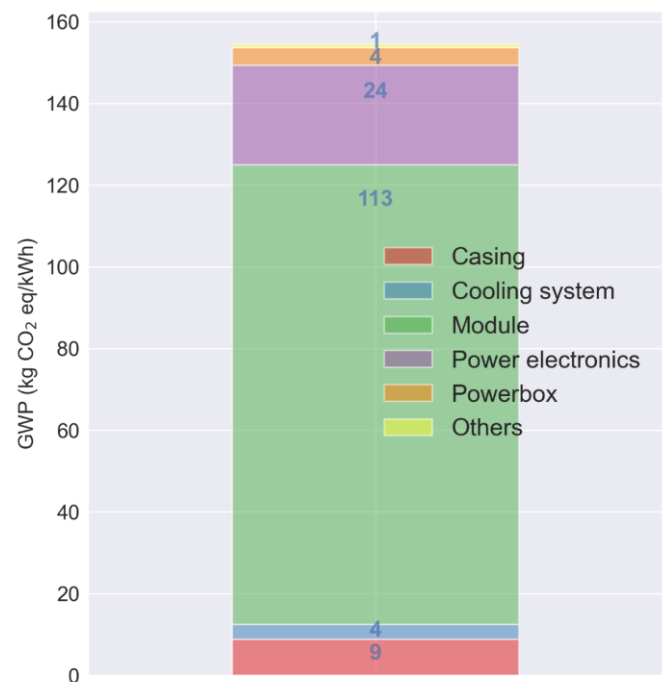


Fig. 5. Environmental impacts for the battery production phase in the climate change impact category with the contribution analysis for the subcomponents

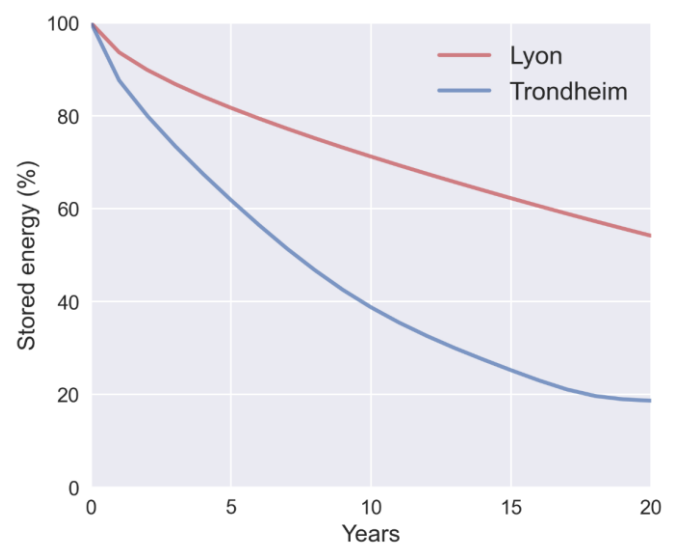


Fig. 6. Capacity loss of a 33.3 kWh (26.6 kWh useable) during 20 years in Lyon and in Trondheim.

In a recent study, Micari et al. [35] assessed the degradation of a LIB under the WLTP class 3 driving cycle. For a cell temperature of 25 °C, for a similar yearly driven distance (16984.18 km), their aged capacity is equal to 74.2% of the initial capacity after 10 years, which is very close to our value. The main difference comes from our electrothermal model that takes into account the variation of the cell temperature depending on the daily external temperatures. As highlighted by Micari et al. [35], this temperature has a strong impact on the loss capacity.

3.3. Comparison of the impact score with and without ageing model

With the ageing model, as explained before, the EoL is reached when the use profile cannot be performed anymore, ie. when the SoC of the battery is equal for the first time to zero. For a 33.3 kWh battery (26.64 kWh useable), the EoL happened at 7 years which leads to a reference flow of 1/7 of battery. As a reference, as mentioned in the introduction, a 10 year is usually arbitrarily chosen for the lifespan, whatever the battery design is. The GHG emissions of the battery under study is 672 kg CO₂eq/year without integrating ageing (lifespan is 10 years) whereas it increases to 959 kg CO₂eq/year when we use the predicted lifespan that comes from our ageing model. This model is very flexible since it can be adapted really quickly for several design configurations or use conditions to recalculate the lifespan. As an example, the external temperature has been changed while keeping the same battery system. The daily temperatures of Trondheim in the year 2020 have been integrated to compute the cell temperature within the ageing model. This has a great impact on consumption (0.2020 kWh/km) and on ageing since capacity losses are higher for low temperatures, as shown in the Fig. 1. Therefore, the predicted lifespan drops to only 3 years in that case which leads to an increase of 233% of the impact as illustrated in Fig. 7. This suggests that benefits from EV could be largely reduced in cold country due to enhanced battery ageing. This conclusion was already introduced by Egede [20] when only looking at consumption and the electricity mix. The ageing mechanism would have been totally ignored by choosing a reference lifespan for every cases, which could lead to wrong decisions when eco-designing a battery system.

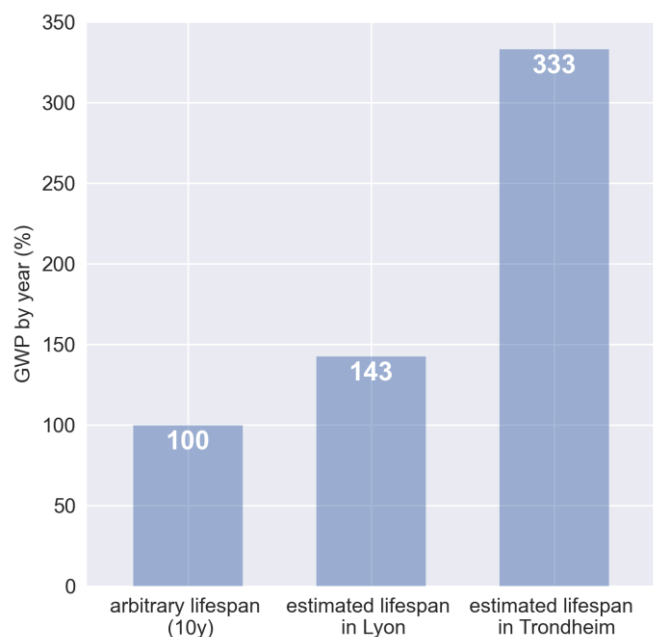


Fig. 7. Annual GWP (normalized) of the studied battery system with and without ageing and using two different daily temperatures in 2020 (Lyon and Trondheim)

4. Conclusion

In this paper, a new approach has been proposed to better predict the lifecycle environmental impacts of Lithium-ion Batteries. A new functional unit has been introduced and an ageing model has been integrated to an LCA in order to predict the lifespan of the battery depending on its design and the use conditions. This model was used to compute the loss capacity of a 33.3 kWh battery from a small-size EV. A lifespan and a number of batteries necessary to fulfill the FU were deduced and permitted to assess the lifecycle GHG emissions of the battery. A significant difference appears when comparing the impact of the studied battery with and without the ageing model. This gap is further enhanced when the battery is used in a cold country, which demonstrates the ability of the model to adapt to several battery design and use conditions. After improving the electrothermal model, this model will be applied on other design parameters in a second time, to perform sensitivity analysis in order to identify what are the main trends and what are the most sensitive parameters when considering the lifespan. This could enable to highlight new environmental hotspots and improve the eco-design of batteries.

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