



Vanadium, molybdenum and nickel: A sustainability analysis of the extraction from ores versus recovery from spent catalysts

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ABSTRACT

The current increasing demand of raw materials has pushed the worldwide research towards the development of innovative processes able to recover metals from end-of-life materials. Considering their content of Mo, Ni and V combined with the availability on the market, the waste hydrodesulfurization catalysts represents an interesting secondary raw material. In the perspective of the implementation of circular economy strategies, a real effective recycling must combine a high efficiency with high environmental sustainability level. With this aim, the present paper carried out a sustainability evaluation by the life cycle assessment (LCA) tool to compare an innovative recycling of catalysts with the primary production of V, Mo, and Ni. The analysis proved that the secondary production allows a CO₂-eq. emission saving higher than 40 %, corresponding to a carbon credit up to around 2,000,000 \$ per year, strongly linked to the kind of supplied energy (country based). The analysis further proved that the waste catalysts, can be integrated with the most common steel scraps treatment with the implementation of a successful and sustainable industry of secondary V.

1. Introduction

Mo, V and Ni are metals extensively used in several fields and are strategic for many of the new technologies required for the future energy transition and sustainability. Access to strategic resources is crucial for the transition to climate neutrality and the sustainability goals claimed in the Agenda 2030 (United Nations, 2024).

Among the three aforementioned metals, V is also included in the fourth critical raw materials (CRMs) list, issued in 2020 by the European Union (EU); such a list reports metals and compounds that are critical to the new European industrial strategy in the main industrial sectors (European Commission, 2020). The European Commission will continue to monitor Ni closely, in view of developments relating to growth in demand for battery raw materials. Although there is not a forthcoming scarcity of Mo and Ni, it has to be pointed out that there are no significant mines and reserves of such metals in the territory of EU-27: this makes EU strongly dependent on foreign countries, in particular those affected by political instability.

In 2022, the primary Mo reserves estimated worldwide were 12 million tons: the most abundant reserves are located in China (3.7

Mtons), followed by USA (2.7 Mtons), Peru (2.4 Mtons), Chile (1.4 Mtons), Russia (430 ktons) and Turkey (360 ktons) (Statista, 2023). In the same year, the top molybdenum producers by country were China (100 ktons), Chile (44 ktons), USA (42 ktons), Peru (32 ktons), Mexico (16 ktons), Armenia (7.8 ktons), Iran (3.5 ktons), Mongolia (2.3 ktons), Russia (1.7 ktons), and Uzbekistan (1.6 ktons) (Investing News, 2023a).

Mo is mostly extracted as molybdenite (MoS₂) with a concentration of around 0.3 %, but also from wulfenite (PbMoO₄) and powellite (CaMoO₄). The co-product molybdenite, which arises from Cu mining, is mainly used for smelting. The MoS₂ concentrate contains around 50–60 % of Mo. The majority of Mo is produced as a by-product in Cu and W extraction and treatment, and only 30 % is extracted from Mo ores. All ores are mainly processed to obtain ammonium heptamolybdate (NH₄)₆Mo₇O₂₄. Thus, molybdenum trioxide (MoO₃) is obtained by roasting that salt at 400–450 °C. Mo powder is another commercial product and can be obtained by reducing MoO₃ by hydrogen in two stages (Institut für Seltene, 2023), Mo is commercialized as Mo metal powder 99.95 %, oxide MoO₃ 57 % or as an alloy, in particular FeMo 65 %.

The majority of Mo is used as active metal in catalysts and in steel

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alloys and super-alloys.

Primary V is extracted from several ore deposits, in particular vanadiferous titanomagnetite, sandstone-hosted (U)-vanadium, calcrete-hosted (U)-vanadium, vanadate, graphite-associated vanadium and vanadium occurrences associated with laterite, bauxite, and phosphate ores (Boni et al., 2023). Secondary V comes from by-products and industrial wastes like oil fly ashes, vanadium bearing steel slag and spent catalysts (Gao et al., 2022).

V is mainly used in steel alloys and super-alloys, as well as catalysts for several processes like reduction of NO_x, sulfuric acid production and cracking of oil feedstock. (Petranikova et al., 2020). In 2022, the estimated world V resources were 63 million tons, whereas the reserves were around 26 million tons; regarding production, the global amount of V was nearly 100,000 tons per year (U.S. Geological Survey, 2023). The production fell to this amount from 150,000 tons in 2021. The leading suppliers were China with 70,000 tons, followed by Russia with 17,000 tons, thus South Africa (9100 tons) and Brazil (6200 tons) (Investing News, 2023b).

The highest Ni reserves in the world are in Australia and Indonesia, with a reserve amount estimated in 21 million tons each, followed by Brazil (16 million tons), Russia (7.5 million tons), New Caledonia (7.1 million tons), Philippines (4.8 million tons), Canada (2.2 million tons), China (2.1 million tons) and USA with 370,000 tons (Investing News, 2023c).

Top Ni producers in 2022 were Indonesia with 1.6 million tons of Ni, followed by Philippines (330 ktons), Russia (220 ktons) and New Caledonia (190 ktons). Australia, despite its huge reserves, was only the fifth world producer with 160 ktons, followed by Canada (130 ktons), China (110 ktons), Brazil 83 ktons and USA, that closed the ranking with 18 ktons (Investing News, 2023c).

Primary Ni is produced from iron ore limonite, which usually contains 1–2 % Ni by weight. Other important Ni ore minerals include pentlandite and Ni-rich natural silicates known as garnierite.

Many of the petrochemical and refinery operations are based on hydrocracking, alkylation, reforming, hydrotreating, and isomerization, that use catalysts based on different active metals (Furimsky, 1996; Kim et al., 2009). In particular, hydrodesulfurization (HDS) makes use of catalysts to produce low-sulfur fuels and meeting pressing environmental regulations (Wang et al., 2021). Catalytic hydrodesulfurization in petroleum refining is carried out with a purpose to eliminate most of the contaminants from liquid petroleum fractions including metals, sulfur, oxygen, and nitrogen. Hydrodesulfurization is carried out prior to catalytic reforming to avoid adulteration of catalysts by the unprocessed feedstock. In addition, hydrodesulfurization is carried out prior to catalytic cracking to improve product yield, reduce sulfur content. These catalysts contain molybdenum sulfide, together with Ni or cobalt sulfide supported by porous alumina (Ruiz et al., 2011). HDS catalysts thus represent valuable secondary reserves for extracting Mo and V, as well as Ni or Co.

Every year, up to 170,000 tons of spent HDS catalysts are generated worldwide (Akciil et al., 2015; Dufresne, 2007). Nevertheless, the current exact amount of recycled catalysts is rather difficult to be quantified, as these confidential data can be obtained from recycling companies only. This huge amount accounts for 4 % of the solid waste produced by refineries (Marafi and Stanislaus, 2008; Shalchian et al., 2019). Spent HDS catalysts are classified as hazardous materials and thus they need special handling and final disposal (Krishnan et al., 2021; Pradhan et al., 2010). High concentration of strategic metals, i.e. V, Mo, Ni or Co makes the HDS catalysts a valuable secondary resource for recycling (Marafi et al., 2017). Recycling of spent catalysts and every kind of industrial waste containing strategic metals is crucial to establish a circular economy approach to increase as much as possible the reuse of natural resources (Ferella et al., 2010, 2018). Moreover, the concentration of certain metals, like those contained in the spent HDS catalysts, is much higher than the concentration they have in primary ores from which they are extracted: this means that their recycling implies a lower

unit energy consumption, minor use of natural resources (rocks and land dug, emissions and water pollution) and a lower environmental impact due to the final landfilling of tailings (Ferella et al., 2019).

The outlook of refining operations in the next year forecasts an increase in the total refining capacity worldwide. The global hydrodesulfurization catalysts market demand was 196.6 ktons in 2019 and is expected to reach 262.7 ktons by 2027 (Grand View Research, 2019).

The advancements in internal combustion engines and increasing regulations concerning environmental pollution are expected to force companies to produce contaminant-free fuels. Owing to these factors, the demand for hydrodesulfurization catalysts is expected to register a significant growth rate over the forecast period, despite the selling of the sole electric cars planned in Europe and California. The rest of the world will continue to produce engines fueled by gasoline and diesel, and this will keep the necessity of refining very high.

Figs. 1 and 2 show the outlook of the HDS global market in terms of quantity and market (Grand View Research, 2019).

The aim of this paper is to compare the environmental impact of the production of V, Mo and Ni from 1) primary ores and 2) from spent HDS catalysts and 3) V from other secondary resources by the life cycle assessment (LCA) tool.

Regarding recycling strategies, different leaching stages can be used to extract metals from HDS catalysts. Water leaching of the catalyst roasted with sodium salt (Huang et al., 2019; Kar et al., 2005), leaching with acids (Barik et al., 2012; Wiecka et al., 2020), alkali leaching (Yaraş and Arslanoğlu, 2020; Zhao et al., 2021) and bioleaching (Pradhan et al., 2013; Srichandan et al., 2015) were used in laboratory and pilot scale studies.

Water leaching is the most used in full scale, as it is the simplest and more effective method requiring no chemical solvents. Nevertheless, the pregnant leaching solution contains different elements, including transition metals, sodium salts, and many impurities such as As and P needing special attention in subsequent separation and purification processes (Wang et al., 2021). The advantage of acid leaching is the highest leaching efficiency, which is obtained by using different organic and inorganic acids (Marafi and Stanislaus, 2008). However, wastewater treatment is the major drawback due to the large quantities of acids used during leaching. The separation of valuable metals such as Mo, V, Ni from the leaching solution of spent catalysts depends on the previous process stages. Precipitation (Park et al., 2006; Pinto and Soares, 2013), solvent extraction (Sahu et al., 2013; Wu et al., 2021), adsorption (Pagnanelli et al., 2011), and ion exchange (Nguyen et al., 2013; Nguyen and Lee, 2014) are frequently used techniques for separating transition metals from leaching solutions. Some spent catalysts are wet, i.e., containing residual naphtha or other hydrocarbon fractions, in a 5–13 % concentration range. Few industrial processes are currently available to recycle HDS catalysts, both pyrometallurgical and hydrometallurgical.

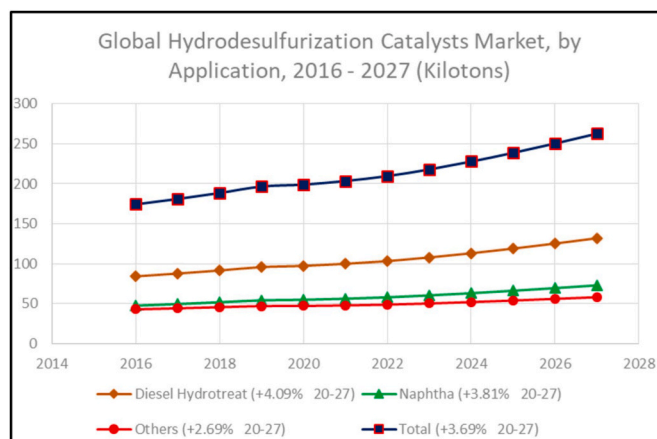


Fig. 1. HDS consumption forecast up to 2027.

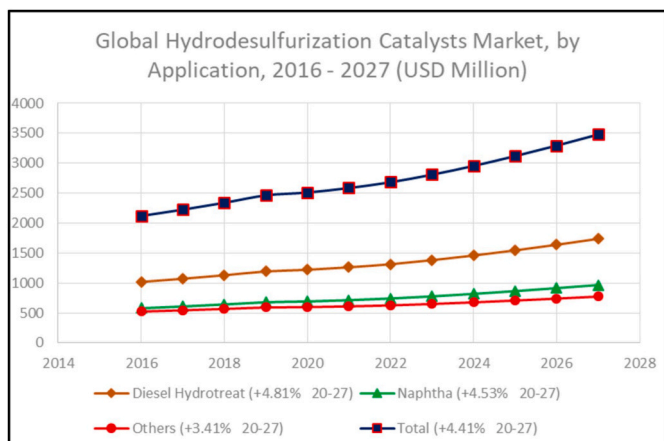


Fig. 2. HDS world catalyst market up to 2027.

Recycling can lead to a circular approach in the oil refining industry, which is the goal of the present study, which proposes a sustainability analysis of a process for the recovery of Mo, V, and Ni from spent HDS catalysts. This paper considered the hydrometallurgical approach, including a double-stage thermal pre-treatment required to oxidize residual hydrocarbons, coke, soot and sulfur.

2. Materials and methods

2.1. Description of the HDS catalyst recycling

The recycling process chosen for the LCA is shown in Fig. 3. This process was described in detail in one of our previous papers, providing energy and material balances (D’Adamo et al., 2023). The capacity of the plant was set at 6000 tons/year of wet Ni-Mo catalyst, working 300 days per year in continuous operation mode. Such catalyst contains nearly 15 %wt of naphtha, 3 %wt each of Mo and Ni, and around 7 %wt of V, the last one coming from the liquid feedstock desulfurized in the refinery. This is a typical concentration of a spent HDS catalyst, but there are many other samples that contains higher concentration of metals, in

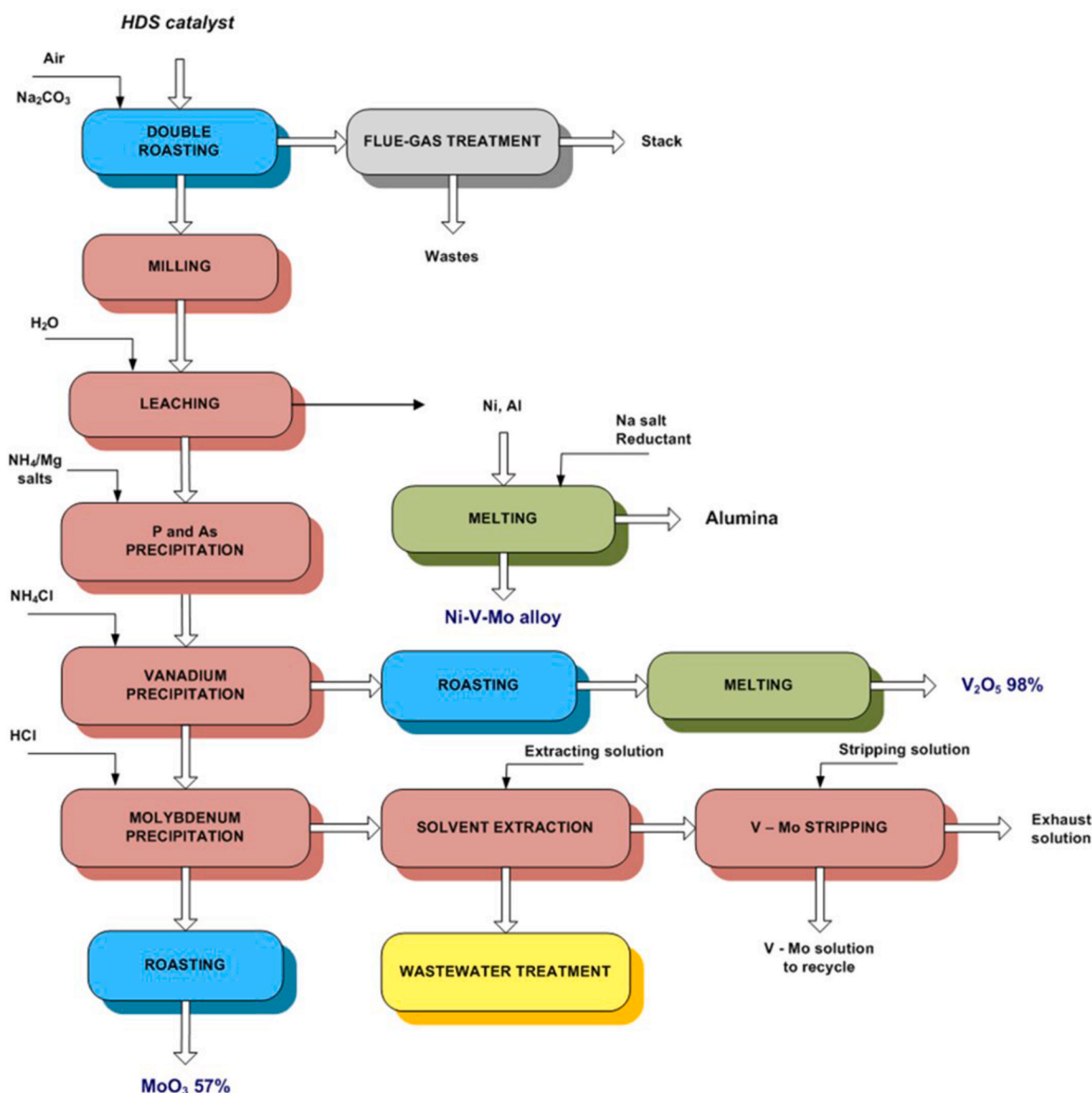


Fig. 3. Flow sheet of the HDS catalyst recycling process (adapted from D’Adamo et al., 2023).

particular Mo (Ferella et al., 2011). Hence, the environmental and economic analysis would be even better than the average case. Naphtha contributes to the total LHV, in addition to sulfur and carbon deposited on the surface, almost eliminating the need for natural gas in the first rotary kiln. The recycling process is divided into five sections: 1) roasting of the spent HDS catalysts, 2) flue gas treatment, 3) hydrometallurgical extraction and recovery; 4) roasting and melting of the products and 5) wastewater and spent solution treatments.

The thermal treatment is carried out in two sequential roasting stages: in the first one, residual hydrocarbons, coke, and sulfur are burnt and Ni, Mo and V are oxidized; in the second stage, Na_2CO_3 is added to let Mo and V convert into sodium molybdate and vanadate, which are water-soluble. Flue gas is treated to remove unburnt compounds, SO_x , and other acidic gases NO_x , and dust. Furthermore, thermal energy is recovered from cooling the flue gas down to produce low-pressure steam, used to heat the reactors of the hydrometallurgical section.

The solid residue from the water leaching stage is washed with water and melted in a EAF to produce an alloy that can be sold to the steel industry.

The pregnant solution is contaminated by arsenic and phosphorous salts that have to be removed as they reduce the quality of Mo and V steel alloys. Thus, ammonium vanadate (NH_4VO_3) and molybdic acid (H_2MoO_4) are recovered by selective precipitation. NH_4VO_3 is decomposed into V_2O_5 and NH_3 , whereas H_2MoO_4 is roasted to obtain MoO_3 . A second EAF melts the V_2O_5 powder into flakes.

All wastewaters and spent solutions are stored in a tank and sent to the wastewater treatment equipment. The treated water is reused in the plant. Hydrometallurgical plants need large amounts of water, so to reduce overall consumption, it is crucial to reuse water and limit the exploitation of groundwater tables.

The plant produces 1272 tons/year of V_2O_5 98 %wt and 239 tons/year of MoO_3 57 %wt, both of commercial grade; moreover, 227 tons/year of Ni/V/Mo alloy with an approximate composition of 72/25/3 % wt are also produced.

2.2. The LCA methodology

The LCA analysis was performed in agreement with the ISO standards 14040 and 14044:2006 ("ISO 14044:2006 Environmental management-Life cycle assessment-Requirements and guidelines," 2006 and UNI EN ISO 14040:2006, Environmental management-life cycle assessment-principles and framework). The software used for data collection is LCA for Experts (by Sphera) version 2024.1, integrated with Professional Database. The method selected for the analyses, which included the classifications and characterizations, normalizations, and weighing steps, was Environmental Footprint (EF) 3.0, and it included all environmental categories and recommended models at midpoint, together with their indicators, units, and sources (Joint Research Center, 2018; Zampori and Pant, 2019). This method was recommended by the European Platform on Life Cycle Assessment as a common way of measuring the environmental performance of processes (European Commission, 2021a, 2021b). The list of the considered impacts, and the related category indexes, can be found in Table S1 in supporting materials.

The functional unit chosen for the present study is the selected plant capacity, i.e. 6000 tons/year of wet Ni-Mo catalyst, working 300 days per year in continuous operation mode. Fig. 4 describes the whole system boundaries considered in the different steps of the presented analysis. Indeed, the first section of the LCA aimed at identifying the main environmental loads and the most affected categories of the innovative process proposed by D'Adamo et al. (2023) for the recovery of V, Mo and Ni from spent HDS catalysts. Input and output flows of the process are summarized in Fig. S1. The analysis compared this scenario with the primary production of the same elements from ores to verify the real advantage of recycling. The further comparison with other processes for metal recovery from steel slugs allowed to estimate the different impacts

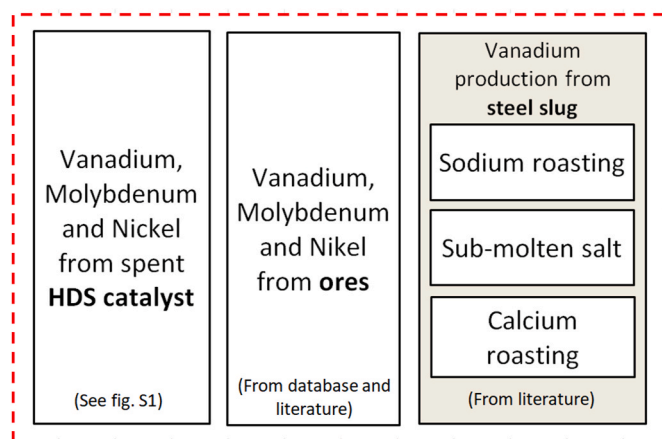


Fig. 4. System boundaries.

due to the V recovery from several kinds of waste.

Table 1 summarizes the inventory analysis necessary for the estimation of the environmental loads, built on the mass and energy balances described by D'Adamo et al. (2023). Some assumptions were performed for the analysis. More in detail, the thermal energy for the two reactors and the multiple evaporator is totally provided by steam generated by the cooling of the fumes of the 4 rotary kilns and the 2 EAFs. The 70 % of the treated wastewater was considered suitable to be recirculated within the same process in the perspective of water cost reduction, since only the 30 % is discharged. These percentages were hypothesized based on the wastewater treatment proposed by D'Adamo et al. (2023). An almost complete recirculation was also assumed for the extractant solution in the solvent extraction step. This choice was performed in agreement with the literature (Amato et al., 2019; Schulze et al., 2017). Furthermore, the extractant amount is lower than 0.07 % of the selected functional unit (6000 tons/year of wet Ni-Mo catalyst) and the assumption cannot significantly affect the whole result. Sodium sulfate from the flue gas treatment was considered as a by-product; therefore, it was considered as a zero-impact flow (more conservative than an environmental credit). The production processes of both raw materials and energy were extracted from the software database, except for ammonium chloride production, not present in dataset and built by synthesis reaction (Nihonium and hydrogen chloride).

As concern the primary production of recovered metals, both Ni and Mo processes were extracted from LCA for expert database, including mining and refining operations. On the other hand, the V impact was estimated from Nuss and Eckelman (2014).

3. Results and discussion

3.1. Sustainability analysis of the HDS catalyst recycling process

The results of classification and characterization allowed the identification of the environmental load of the process steps on each impact category considered for the analysis.

Fig. 5 reports the contribution of each stage on the whole process burden, highlighting the main issues of the flue gas treatment, the melting of leaching residue and the utility supply, due to their high-energy demand. This aspect is especially enhanced in the categories of ionizing radiation, land use and ozone depletion. The result is strongly linked to the specific grid mix chosen for the analysis, i.e. the average EU electricity mix. For example, the effect on ionizing radiation-human health is due to the radionuclides (potentially toxic for humans) resulting from both the nuclear energy production, and the mineral oil and gas extraction, used as energy carriers (Amato et al., 2019; Çankaya and Pekey, 2020; Frischknecht et al., 2007). The effect of grid mix change on the whole environmental load was discussed in the sensitivity

Table 1
Input and Output of the process.

Operation	Input	Output	Operation	Input	Output
Double-stage roasting (2 kilns)			Grinding		
Methane (10 ³ Nm ³ /y)	398		Electrical energy (MWh/y)	216	
Sodium carbonate (t/y)	1080		Waste dust (t/y)		10
Electrical energy (MWh/y)	900				
Flue gas treatment			Purification of solution		
Electrical energy (MWh/y)	576		Electrical energy (MWh/y)	104	
Sodium carbonate (t/y)	1404		MgO solid (t/y)	35	
Ammonia (t/y)	58		NH ₄ Cl solid (t/y)	41	
NaOH solid (t/y)	288		Water (t/y)	139	
Waste Na ₂ SO ₃ (t/y)		403	HCl 37 % liquid (t/y)	72	
Waste Na ₂ SO ₄ (t/y)		1872	NaOH solid (t/y)	2	
			Wash wastewater (t/h)		166
			Waste MAP-MAAs sludge (t/y)		144
Roasting of AMV			Leaching stage		
Methane (10 ³ Nm ³ /y)	193		Electrical energy (MWh/y)	157	
Electrical energy (MWh/y)	83		Water (t/y)	16,481	
V ₂ O ₅ 98 %min (t/y)		1272	Wash wastewater (t/y)		3521
Vanadium precipitation			Melting of leaching residue		
Electrical energy (MWh/y)	396		Electrical energy (MWh/y)	12,167	
HCl 37 % liquid (t/y)	36		NaOH solid (t/y)	108	
NH ₄ Cl solid (t/y)	778		Coal (t/y)	43	
Water (t/y)	2606		Ni/V/Mo alloy (t/y)		227
Wash wastewater (t/y)		2290	Waste alumina slag (t/y)		2196
Molybdenum precipitation			V-Mo stripping		
Electrical energy (MWh/y)	97		Electrical energy (MWh/y)	94	
Reductant (t/y)	31		NaOH solid (t/y)	14	
HCl 37 % liquid (t/y)	432		Water (t/y)	7214	
Wash wastewater (t/y)		374	NaCl solid (t/y)	43	
Solvent extraction			Wastewater treatment		
Electrical energy (MWh/y)	230		Electrical energy (MWh/y)	221	
H ₂ O ₂ 30 % liq (t/y)	26		Wastewater released (t/y)		9154
Kerosene (t/h)	108		Wastewater recycled (t/y)		21,360
Extractant (t/h)	4		CaO (t/y)	155	
Waste solution (t/y)		23,760	H ₂ SO ₄ 98 % (t/y)	18	
			FeSO ₄ *7H ₂ O (t/y)	12	
			H ₂ O ₂ 30 % liq (t/y)	71	
			Waste sludge (t/y)		180

Table 1 (continued)

Operation	Input	Output	Operation	Input	Output
Roasting of amoly			Melting of V₂O₅ powder		
Methane (10 ³ Nm ³ /y)	107		Electrical energy (MWh/y)	4117	
Electrical energy (MWh/y)	156		V ₂ O ₅ flakes 98 % min (t/y)		1272
MoO ₃ 57 %min (t/y)		239			
Storage tanks and reagent dilution			Utilities		
Electrical energy (MWh/y)	354		Methane (Nm ³ /y)	140	
			Electrical energy (MWh/y)	5986	
			Cooling water (t/y)	38,232	

analysis section.

The use of huge quantities of sodium carbonate, produced by Solvay (ammonia-soda) process, justify the impact of the double-stage roasting and at least 35 % of the flue gas treatment, in most of environmental aspects.

A completely different trend was observed for the human health effects (both cancer and non-cancer), where more than 95 % of the environmental burden is due to the disposal of MAP-MAAs sludge, classified as hazardous waste.

Table S2 reports the impact values for all the categories included in the study.

The results of classification and characterization highlighted the main environmental issues of the process. However, the results are not completely homogeneous for all the categories. The further normalization and weighting allowed to compare the different categories by using normalization and weighting factors obtaining a whole result, expressed as person equivalent, i.e., the number of people (average European citizens) that generates the same effect per year (Schmidt and Frydenal, 2003). The achieved result, reported in Fig. 6, identifies as the greatest environmental issues the climate change and the resource use fossils, due to the electricity demand. All the steps of the treatment contribute to both these categories with a different load, as showed in Fig. 7. The ecotoxicity contribution (around 20 %), is linked to the production of sodium carbonate used for the double-stage roasting and the flue gas treatment (Fig. 6).

3.2. Recycling vs. primary production, the effect on climate change

The supply of raw materials represents a hot topic in Europe, currently interested in the identification of new metal resources. This interest is confirmed by the inclusion of V and Ni in the critical raw materials list of 2023. However, in the perspective of the reduction of climate change effect the only identification of new resources is not enough; the innovative processes should be more sustainable than the traditional one, with resulting positive effect for the environment. To verify the sustainability of the proposed technology, its effect on climate change was compared to the traditional ways for the primary production of V, Mo and Ni. The results in Fig. 8 show a potential benefit of 44 % resulting from the substitution of primary production from ores, with the innovative HDS catalyst recycling process. The results highlight that the greatest gain is due to the avoided primary production of V due to both the highest recovered quantity (769 tons for year, in the considered plant) and the greatest impact per kg of metal. Indeed, the kg of CO₂ eq. produced for the traditional extraction and refining per kg of V are around 5 times higher than those estimated for Ni and Mo.

To better understand the magnitude of this environmental gain, the avoided 12,480 tons CO₂ eq. are comparable to the average carbon footprint produced by 1545 European people and 1846 worldwide

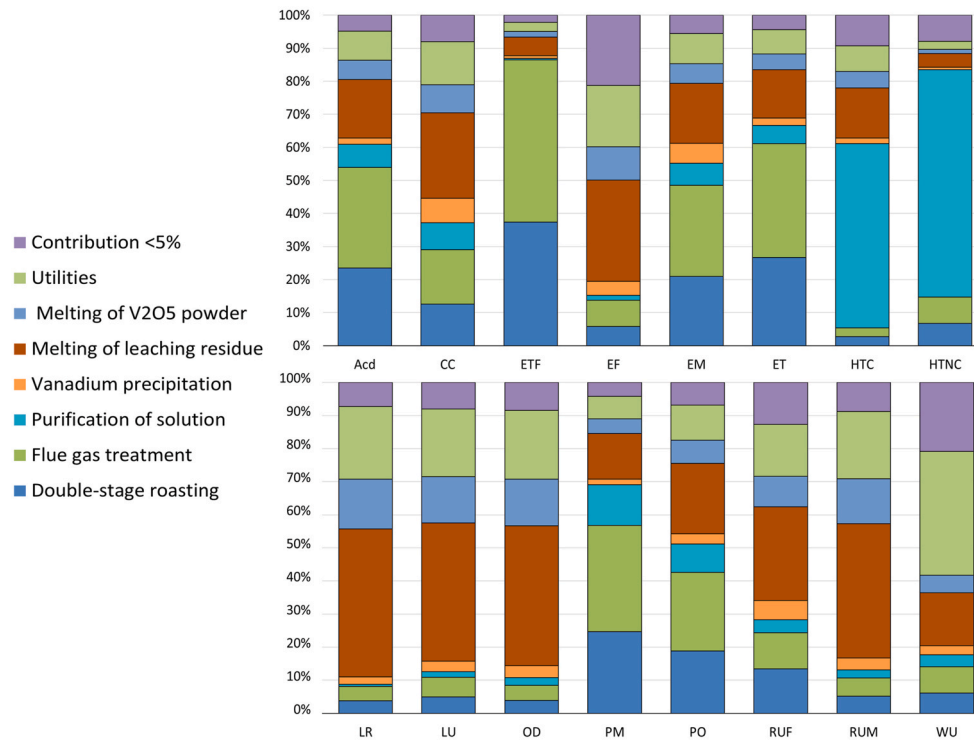


Fig. 5. Classification and Characterization results quantified with the method environmental footprint 3.0. (Acid- Acidification; CC-Climate Change; ETF-Ecotoxicity, freshwater; EF-Eutrophication, freshwater; EM-Eutrophication, marine; ET-Eutrophication, terrestrial; HTC-Human toxicity, cancer; HTNC-Human toxicity, non-cancer; LR-Lonising radiation, human health; LU-Land Use; OD-Ozone depletion; PM-Particulate matter; PO-Photochemical ozone formation; RUF-Resource use, fossils; RUM-Resource use, mineral and metals; WU-Water use).

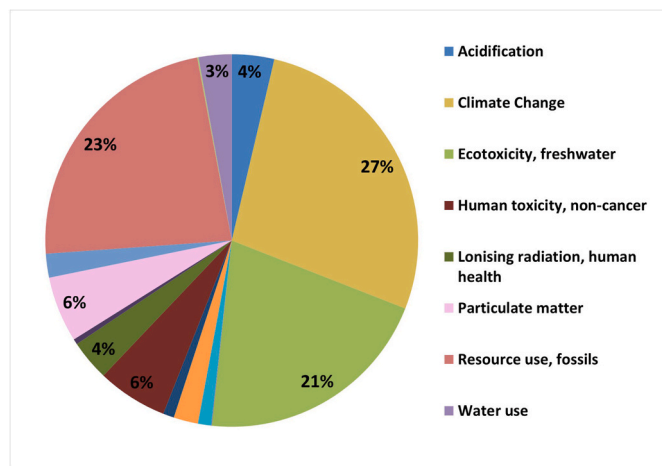


Fig. 6. Normalization and weighting results. (Results with a contribution <3 % are not present in the legend).

(considering respectively 8.08 and 6.76 ton of CO₂ pro capita in 2022 (Martin et al., 2023)). In agreement with the most recent World Bank Group guidelines, showed in Fig. 9, the economic value of the environmental credit resulting from the HDS recycling process (referred to 5985 ton/y used HDS catalysts) is estimated between 5741\$ and 2,084, 430\$, the gap depend on local legislation and it is represented by Carbon Tax or ETS for the carbon credit market. The lowest price is the Indonesian one (0.61\$/ton CO₂) while highest is that of Uruguay (167.17 \$/ton CO₂). The highest ETS are the European estimated in 61.3 \$/ton CO₂ (765,123\$) (World Bank Group, 2024).

3.3. Sensitivity analysis, the effect of electricity grid mix

The LCA of the HDS recycling process identified the energy consumption as one of the most relevant issues. Nevertheless, the environmental load due to the electricity production can change with the specific grid mix, selected on the basis of the country where the plant is located. The results showed in Sections 3.1 and 2.2 referred to the average European grid mix, where nuclear and natural gas are the greatest energy resources (Fig. 10). Nevertheless, considering the worldwide availability of spent catalysts and the possibility to implement this kind of plants everywhere, a sensitivity analysis was performed to study how the selected grid mix can affect the whole environmental impact of the innovative recycling. With this aim, three countries with different grid mix were selected, China and USA considered as the major manufacturing and commercial powers and Italy, with a natural gas contribution of 47 % of the production and the only one (among the considered countries) with a significant component of solar source.

The results described in Fig. 11 show that the specific grid mix can significantly affect the process impact on climate change. Overall, the recycling process is confirmed as the best choice (compared to the primary production), with the exception of the Chinese scenario, where the recycling result is comparable to that of mining. This achievement is justified by the major contribution of hard coal (more than 60 % of the Chinese electricity grid mix), well known for its mammoth contribution on climate change (Edwards, 2019). The same reason (20 % of the American energy is produced by hard coal resources) justifies the impact increase if the process is implemented in USA.

On the other hand, European mix shows the best result due to the use energy resources with low climate change emissions (e.g. nuclear power and hydroelectric). These results suggest that, in the perspective of a further process scale-up, the choice of the kind of energy to supply plays an essential role for the sustainability. In the Italian scenario, where a

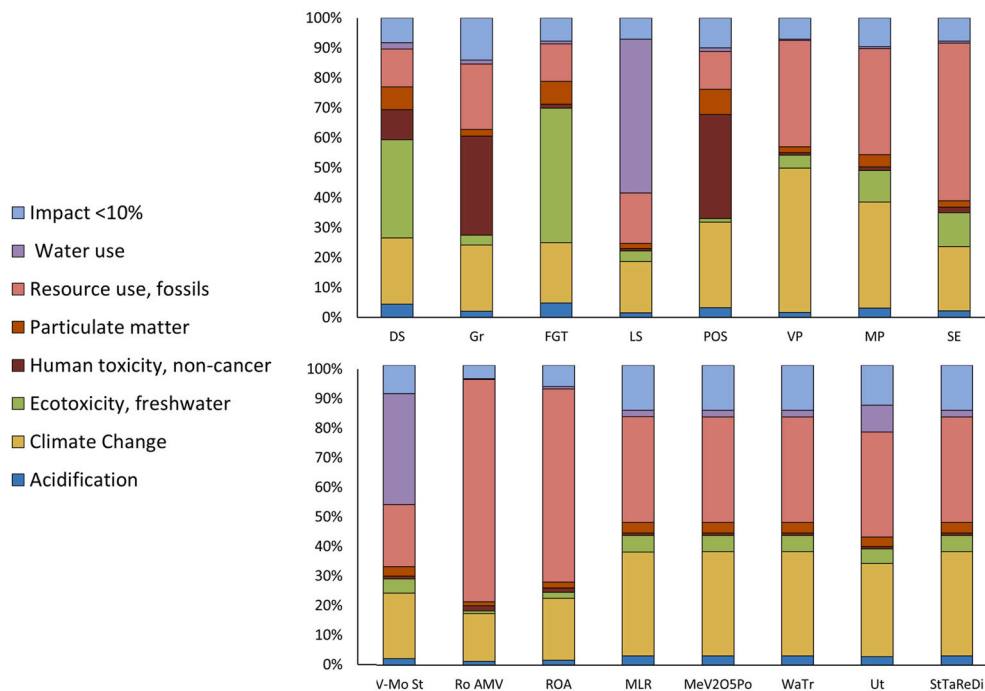


Fig. 7. Normalization and weighing results. (DS-Double stage roasting; Gr-Grinding; FGT-Flue gas treatment; LS-Leaching stage; POS-Purification of solution; VP-Vanadium precipitation; MP-Molybdenum precipitation; SE-Solvent extraction; V-Mo St-V-Mo stripping; Ro AMV-Roasting of AMV; ROA-Roasting of amoly; MLR-Melting of leaching residue; MeV2O5Po- Melting of V2O5 powder; WaTr-Wastewater treatment).

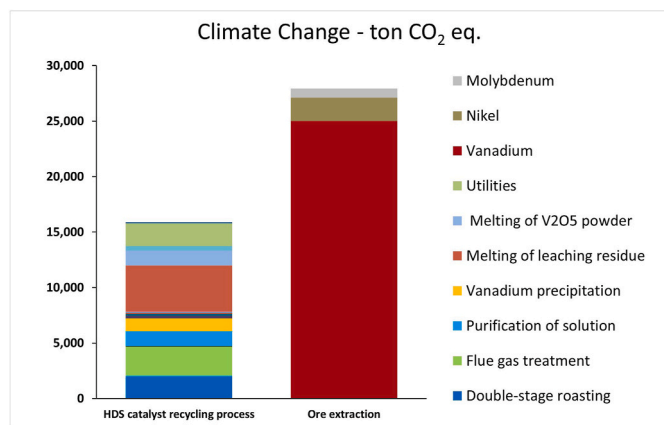


Fig. 8. Climate change category impact comparison (functional unit: 6000 tons/year of wet Ni-Mo catalyst). Results <10 % are not present in the legend.

solar contribution was already included, the further integration of the recycling process with a solar panels system could decrease the impact due to the energy production up to 90 % (0.393 kg CO₂ eq./kWh from energy grid mix, vs. 0.026 kg CO₂ eq./kWh from solar energy).

3.4. Comparison of V recovery from HDS catalysts with other secondary resources

The current availability of spent catalyst is around 170,000 ton/year (Akcil et al., 2015; Dufresne, 2007). Considering an average conc. of V in the spent catalysts around 13 % and a metal consumption around 100, 000 ton/year of V per year (U.S. Geological Survey, 2023) and high-efficiency recycling processes (as that described in the present paper) could cover up to 20 % of the whole demand. Additional secondary resources must be identified to cover a greater fraction of V demand. In this regard, the literature reports several processes for the treatment of V-rich by-products and industrial waste (Zhang et al.,

2023). Considering the sustainability pillar for the development of an effective circular economy, Fig. 12 compares the kg CO₂ eq./kg of recovered V resulting from HDS catalysts with other recycling processes from the literature. In this regard, Zhang et al. (2023) describe three processes of V production from steel slug: Sodium roasting process, Calcium roasting and sub-molten salt.

The sustainability of HDS catalysts recycling is confirmed compared to both the sodium roasting, which represent the most common (over 90 %) secondary production of V from slag, and the sub-molten salt. On the other hand, calcium roasting shows a carbon footprint comparable to the catalyst recycling, lower than the primary production of V.

3.5. Innovation of the study, comparison with other sustainability analysis in the literature

The current attention for the identification of sustainable strategies for V recovery has pushed the scientific community to conduct studies about this topic. Baritto et al. (2024) describe a process for the metal recovery from bitumen upgrader spent catalysts, only focusing on the category on climate change (Baritto et al., 2024). The result, only in the considered category, is comparable with that achieved in the present paper (2.7 kg CO₂/kg spent catalyst vs 2.3 kg CO₂/kg estimated by Baritto et al. (2024)). Several categories were assessed in other papers, which considered different secondary raw materials, such as vanadium titanomagnetite, slags and stone coal (Chen et al., 2015; Jia et al., 2014; Zhang et al., 2023). Shakibania et al. (2024) describe an interesting study for V recovery from spent catalysts including the steps of leaching, purification, precipitation, and calcination. The LCA carried out by the authors mainly aims at the identification of the process weaknesses and excludes the further recovery of valuable elements such as Ni and Mo (Shakibania et al., 2024). The catalyst recycling treatments are also discussed from an economic point of view, emphasizing the profitability of these processes, also considering the current V market price (Baritto et al., 2022; D'Adamo et al., 2023). In this context, the innovation of the present analysis relies on the description of the environmental sustainability aspects of a recycling process implement in an actual industrial

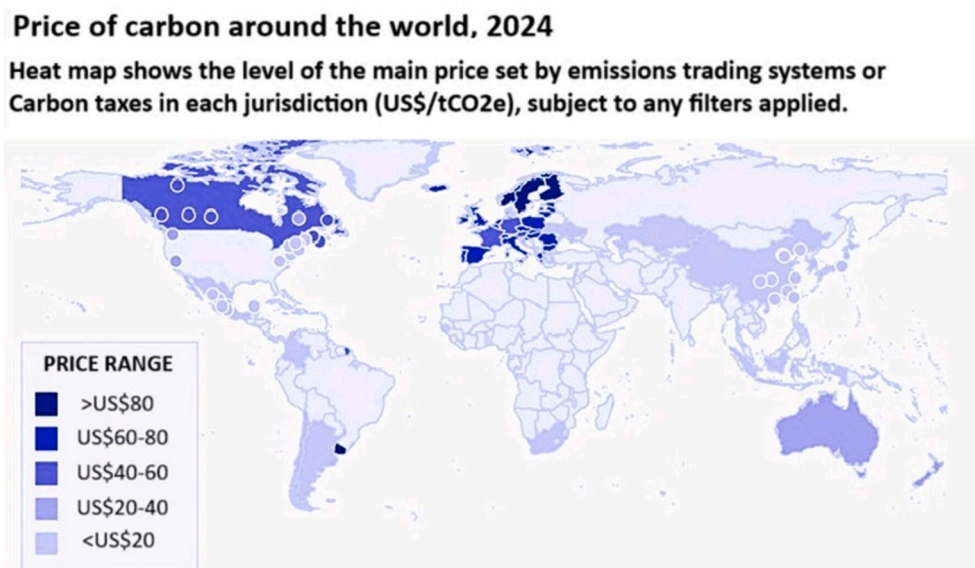


Fig. 9. Price of Carbon credit around the world- Figure adapted from World Bank Group (World Bank Group, 2024).

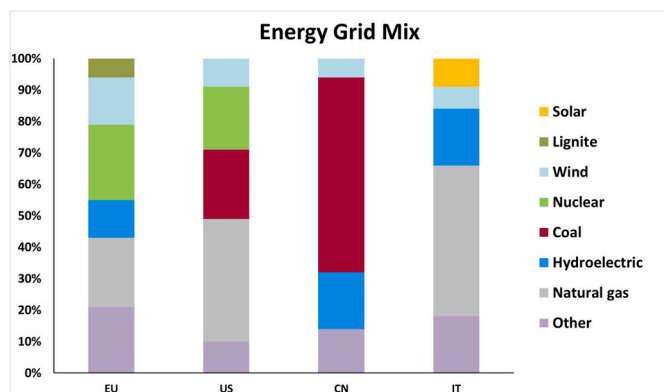


Fig. 10. Description of the different electricity grid mix on the country basis.

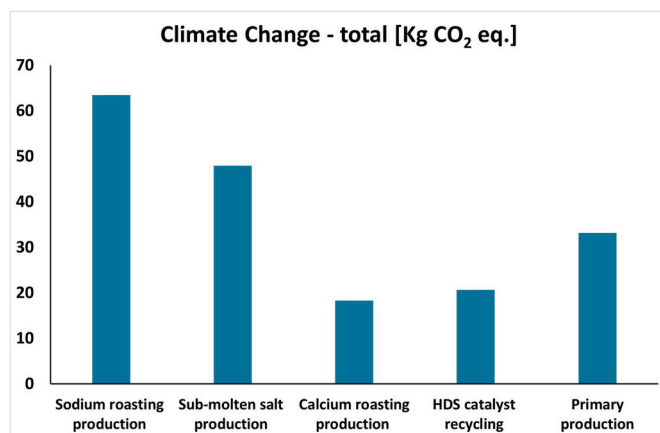


Fig. 12. Comparison with other vanadium secondary productions from the literature (adapted from Zhang et al., 2023).

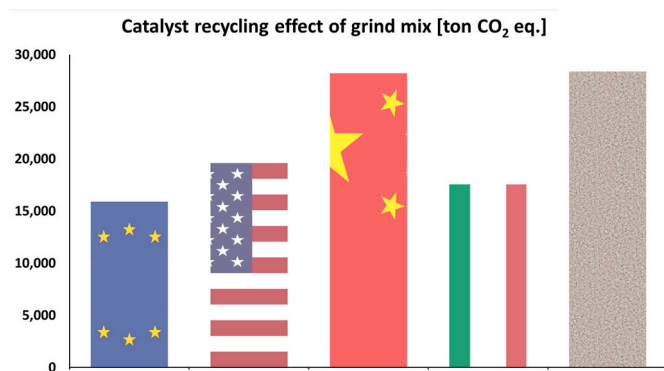


Fig. 11. Comparison among the estimated impacts on the selected country (and energy mix) basis (referred to 6000 t/y used HDS catalysts).

plant (described by D’Adamo et al. (2023)), including all the categories identified by the most updated and credible method identified by the European Commission (EF 3.0). The further comparison with the most common V, Mo and Ni from ores (that still lacked in the literature about catalyst enhancement), made credible by a robust sensibility analysis, represents an important food for thought for the recycling industry.

4. Conclusions

The creation of a secure supply chain of metals is necessary to support the current green transition and an efficient industrial growth. The present analysis considered an innovative process for the recycling of V, Mo and Ni from spent catalyst proving that a 44 % CO₂-eq. emission saving is possible, compared to the primary production from ores. This value can be also translated into an economic income, estimated by the carbon credit method, able to reach up to 2,084,430\$ per year. The LCA analysis allowed to identify the energy consumption as the main issue of the treatment, mainly due to the steps of gas treatment, melting of leaching residue and utility supply (with an average contribution of 70 % on the whole impact, irrespective of the selected category), proving the relevance of the specific grid mix of country where the process is implemented. In this regard, a high contribution of hard coal can significantly decrease the sustainability of the treatment, making it comparable to the traditional mining and refining. The results demonstrate that the transition from a linear economy to a sustainable circular economy is possible by the combination of technological innovation and environmental sustainability tools.

CRediT authorship contribution statement

Alessia Amato: Writing – original draft, Validation, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Nicolò M. Ippolito:** Investigation. **Matteo D’Arcangelo:** Writing – review & editing, Writing – original draft, Visualization, Software, Investigation, Formal analysis. **Alessandro Becci:** Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Formal analysis. **Valentina Innocenzi:** Resources, Funding acquisition. **Francesco Ferella:** Supervision, Resources, Project administration, Funding acquisition, Data curation, Conceptualization.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jclepro.2025.145817>.

Data availability

Data will be made available on request.

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