

**Preliminary report****Environmental assessment of copper slag aggregate concrete**Sandra Filipović^{*1)}, Snezana Marinković²⁾, Dimitrije Zakić²⁾¹⁾ Mining and Metallurgy Institute Bor, 19210 Bor, Serbia²⁾ University of Belgrade, Faculty of Civil Engineering, Department for Materials and Structures, Bulevar kralja Aleksandra 73, 11000 Belgrade, Serbia**Article history**

Received: 26 April 2022

Received in revised form:
19 May 2022

Accepted: 27 May 2022

Available online: 30 June 2022

Keywordsconcrete,
copper slag aggregate,
Life Cycle Assessment,
environmental impact**ABSTRACT**

Due to the extremely large global production and utilization of concrete, the concrete industry is considered a large consumer of energy and natural resources and one of the main sources of greenhouse emissions and waste generation. Reducing its impacts on the environment is one of the most important paths toward sustainable construction development. Replacing cement and natural aggregates with by-products and waste from other industries is one possible way of achieving this goal. On the other hand, the disposal of industrial waste, which arises from the pyrometallurgical process of copper production, presents a significant environmental load in many developed countries. Research performed so far has shown that concrete made with aggregate obtained from copper slag instead of natural aggregate can be produced with good physical-mechanical properties for various applications. In this work, a cradle-to-gate Life Cycle Assessment of several concrete mixtures where part of the coarse natural aggregate was replaced with copper slag aggregate was performed. The conducted case study was based on Serbian LCI data and local conditions in the vicinity of the town of Bor in Serbia. Results showed that such concrete mixtures can bring environmental benefits regarding natural aggregate preservation and waste reduction if the transport distance of copper slag aggregates is smaller than 20 km. Therefore, from the environmental protection point-of-view, local application of such concrete is recommended.

1 Introduction

Concrete is the most commonly used construction material in the world with an average annual global production of about three tons per capita, or a total average production of about 25×10^9 tons per year [1], due to its durability and great possibilities of application, simplicity of production, and availability of raw materials [2]. Aggregates occupy between 55% - 80% of the volume of concrete [3]. Natural aggregates (NA) refer to the product of crushed stone produced in quarries, or extracted gravel and sand from the riverbed. However, many countries have a problem with a deficiency of quality sand on the one hand, and growing environmental problems on the other. An example of this is the fact that the current rate of extraction of NA can cover only 9% of the total annual needs in China [4]. On the other hand, the production of primary products in different industrial activities results in various by-products that have almost no practical industrial application. These industrial by-products, which are generated in high quantities worldwide, present serious challenges regarding their disposal [5]. As a result, more ecologically suitable alternatives for their utilization must be found.

Industrial waste, which arises from the pyrometallurgical process of copper production, is one of the most significant

environmental problems in many developed countries. It is estimated that for every ton of produced copper, about 2.2 - 3.0 tons of slag are generated. Approximately 40 million tons of slag are generated from copper production in the world each year [6]. The frightening data on the annual volume of dumped waste material is cause for worldwide worry, and a solution to the problem must be required in accordance with generally acknowledged principles of sustainable development. One possible solution is the use of copper slag aggregates (CSA) as a replacement for NA in concrete [7].

In pyrometallurgical copper processing, different types of slag are produced: smelter slag, converter slag, anode furnace slag, electric and flash furnace slag, etc. Most of these slags, especially those with higher copper content, are recycled by re-entering the process. The concentration of copper in smelter slag usually ranges from 0.5-2.0% (0.3-1.0% from flame furnaces and over 1% from the flash furnace), while converter slag can contain 2-10% of copper.

Slag with copper ore content below 1% has traditionally been considered solid waste and has been disposed of at landfill. This kind of slag has become a significant secondary raw material [8]. One solution to the problems of industrial waste disposal is the application of copper slag in construction. Due to favorable physical-mechanical and chemical characteristics of copper slag, such as low water

* Corresponding author:

E-mail address: filipovic@irnbor.co.rs

absorption (water absorption of aggregate (WA_{24}) 0.6%), high particle density (apparent particle density (ρ_{rd}) 3.33 Mg/m^3), good resistance to crushing (resistance to degradation (LA) 10%) and wear (resistance to wear (micro-Deval) in a wet state (M_{DE}) 4%), its use is possible in the production of building materials [9]. Copper slag in building materials has the potential to increase mechanical properties while also lowering costs and easing disposal issues [10].

A large number of studies have been performed on the application of copper slag in concrete: as a substitute for cement, i.e. in the production of cement clinker [11, 12], and as partial or complete substitutes for coarse and fine fractions of NA in concrete mixtures and cement mortars [13, 14]. Previous studies have shown that around 40% replacement of NA with CSA gives a concrete mixture with better properties such as compressive strength, density, workability, durability, or flexural strength in concrete [15]. Tests have shown, for instance, that the optimal use of CSA for replacing fine fractions of NA (grains 0-2 mm) is 40% [16]. Such concretes have good workability, and higher compressive and tensile strengths, but due to the lower water absorption of the aggregate from copper slag, care must be taken when choosing a water-to-cement ratio (w/c) [17]. The lower number of tests was related to the physical-mechanical properties of concrete with partial replacement of coarse fractions of NA with CSA [18, 19]. Insufficient results of research into the material itself is the reason for the poor use of concrete with CSA as a substitute for coarse fractions of NA. It could give a clearer picture of the mechanism that takes place during the hardening process and enable the development of generally accepted design principles.

The industrial processing of smelting slag within the Mining and Smelting Basin Bor (RTB "Bor") has been carried out in the Bor Flotation Plant since 2001. Copper slag has been deposited in the immediate vicinity of processing facilities, in the amount of 16 – 18 x 10⁶ t, which was the incentive for testing the possibility of replacing NA with CSA in concrete. The process for obtaining the copper slag generally consists of two-stage crushing and separation according to Figure 1. Due to its high hardness, crushing of copper slag results in high electricity consumption and high damage to the crushers used in this process. For that

reason, naturally granulated copper slag from that landfill was used for testing. Considering that in the natural deposited state of copper slag at RTB "Bor", about 70% of deposited material belongs to the 8/16 and 16 / 31.5 mm fractions, testing was performed on concrete mixtures where part of these fractions of NA was replaced with CSA.

2 Mechanical and durability related properties of CSAC – own investigation

Concrete with a replacement of coarse NA with CSA is designated in this work as concrete with copper slag aggregate (CSAC). Within our own investigation, CSAC with two replacement ratios of NA was tested: CSAC_20_50 concrete in which 20% of 8/16 mm fraction and 50% of 16/31.5 mm fraction were replaced by volume with corresponding fractions of CSA, and CSAC_50_50 concrete in which 50% of 8/16 mm fraction and 50% of 16/31.5 mm fraction were replaced by volume with corresponding fractions of CSA. A control mix of concrete with only NA (NAC) was also tested. Standard methods used for the mix design and testing of NAC concrete can also be used for the design and testing of CSAC mixtures. Mix proportions of concrete together with tested compressive strength at 28 days and flow table results are presented in Table 1 [9]. The control concrete mixture was designed to meet the requirements for the C25/30 strength class.

The densities of fresh concrete mixtures CSAC_20_50 and CSAC_50_50 were higher by 4% and 5%, respectively, compared to the NAC. The density of hardened concrete mixtures CSAC_20_50 and CSAC_50_50 was higher by 5% and 7%, respectively, compared to the control mixture. This was due to the higher density of CSA grains compared to the NA grains. As shown in Table 1, mixtures CSAC_20_50 and CSAC_50_50 showed slightly larger compressive strengths at 28 days compared to NAC, the increase being equal to 12.4% and 10.5%, respectively. The cause of this increase was the higher compressive strength of CSA grains and better interaction between grains, due to their roughness and sharp edges.

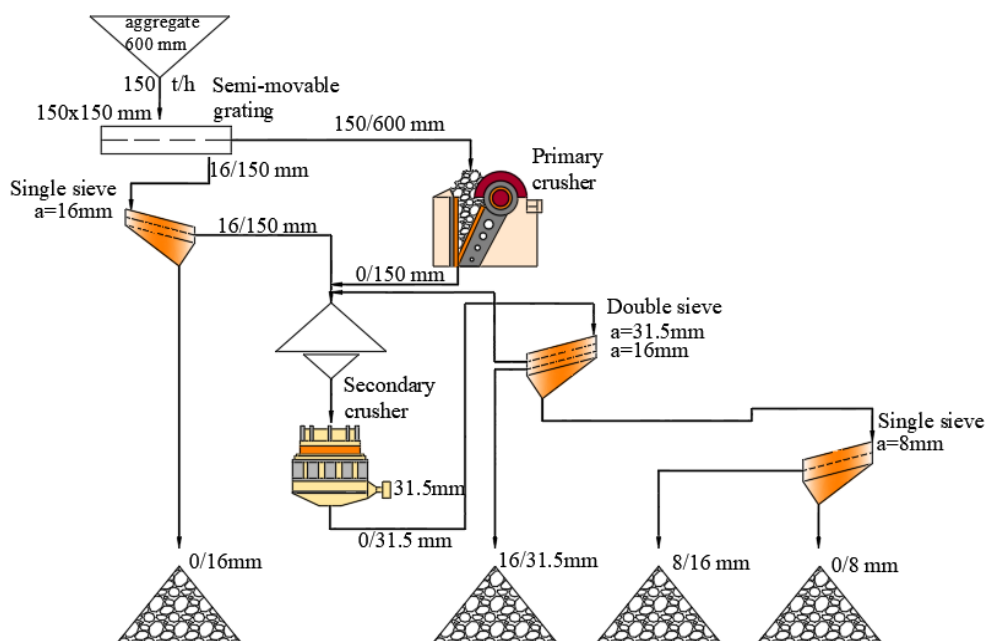


Figure 1. Copper slag plant technology

Table 1. Mix proportions, flow and compressive strength of tested NAC and CSAC [9]

Type of concrete	Cement (kg/m ³)	Water (kg/m ³)	Aggregate			w/c /	Flow (cm)	Compress. strength 28 days (MPa)
			Fine NA (river) (kg/m ³)	Coarse NA (river) (kg/m ³)	CSA (kg/m ³)			
			NAC	398	212			
CSAC_20_50	398	212	732	753	394	0.533	32.0	40.8
CSAC_50_50	398	212	732	659	529	0.533	28.5	40.1

However, the replacement of NA with CSA caused the stiffer consistency of CSAC mixtures compared to the control mixture, as shown in Table 1. This occurred because the CSA grains had a larger specific surface area and less mobility. CSA grains are rougher and have sharper edges than NA grains, which causes greater friction between grains.

The water penetration resistance of the mixtures CSAC_20_50 and CSAC_50_50 was lower by 55.4% and 27.7%, respectively, compared to the NAC. At first sight, the result obtained in this way did not seem logical since the CSA itself had lower water absorption than NA. However, the water penetration resistance of concrete depends on the compactness of the concrete mixture, i.e., the presence of capillary pores in the cement stone. The increase in the volume of capillary pores in the concrete mixtures directly causes the decrease in the mixtures' water resistance. Although the same w/c ratio was used in all mixtures, due to the lower water absorption of CSA grains, the content of free water in the mixtures CSAC_20_50 and CSAC_50_50 was increased. The volume of capillary holes in concrete mixtures increased in direct proportion to the rise in free water.

Also, the increase in chloride penetration classified by the D_{RCM} chloride migration coefficient was observed in the CSAC_20_50 mixture. The recorded effective chloride migration coefficient D_{RCM} for the NAC showed a value of 39.7 x 10⁻¹² cm²/m, while for the CSAC_20_50 and CSAC_50_50 mixtures, D_{RCM} was 46.8 x 10⁻¹² cm²/m and 36.0 x 10⁻¹² cm²/m, respectively. With a D_{RCM} value higher than 16 x 10⁻¹², all three mixtures could be suitable for use in environments without aggressive influences [20, 21]. However, as with water penetration resistance, the main

pattern of enhanced chloride migration through concrete is due to the increasing porosity of cement stone.

3 Environmental assessment

Environmental assessment was conducted using a well-recognized and standardized methodology for evaluating the environmental loads of processes and products during their life cycle - Life Cycle Assessment (LCA). According to ISO 14040 standards [22], LCA consists of four steps: (1) goal and scope definition, (2) creating the life cycle inventory (LCI), (3) assessing the environmental impacts (LCIA) and (4) interpreting the results.

3.1 Goal, system boundaries, and functional unit (FU)

The goal of this study is to assess the environmental potential of replacing the NA with the CSA in the production of ready-mixed concrete. In the basic scenario, natural river aggregate was chosen instead of crushed aggregate since it is commonly used in Serbia. Within the CSAC mix design, two different replacement percentages of NA with CSA were tested: CSAC_20_50 and CSAC_50_50. Table 1 shows the mix proportions of the alternatives with the evaluated compressive strength after 28 days and the flow table results.

Having in mind this goal, a cradle-to-gate analysis was conducted, Figure 2, and the functional unit of 1 m³ of ready-mixed NAC and CSAC was used in this work. The production of water was excluded from the analysis.

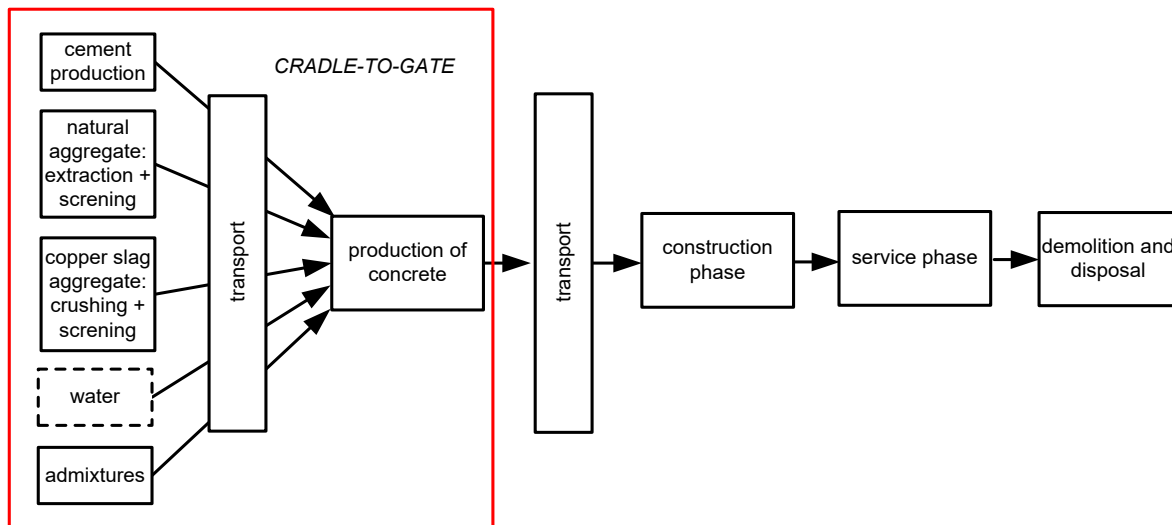


Figure 2. Cradle-to-gate part of the CSAC life cycle

3.2 Life cycle inventory (LCI) and transportation data

The production of ready-mixed NAC and CSAC studied in this paper is located in Bor, Serbia. All the LCI data for natural river aggregate, cement and concrete production was collected from local suppliers and manufacturers [23]. As for the CSA, data was collected from the flotation plant of the Mining and Smelting Basin Bor, where the copper slag is deposited at the landfill near the processing facilities. Since only naturally granulated copper slag was used in this research, the process for obtaining the copper slag consisted of sieving according to Figure 3. The electricity consumption is 0.28 kWh (1.01 MJ), while for the internal services, 0.704 l of diesel (24.8 MJ) is used per tone of aggregate. Emission data for diesel production and distribution, as well as for transportation that could not be collected for local conditions, was taken from the Ecoinvent V2.0 database [24, 25].

According to European Directive 2008/98/EC [26] any product that can bring revenue should not be considered as merely waste but as a useful by-product. This means that it carries a part of the environmental load of the primary process (copper production - main product), besides the load from its own treatment prior to utilization in concrete (secondary process – by-product). In the case of the application of various by-products or wastes as mineral additions or cement replacements in concrete, economic allocation is usually recommended over mass allocation. Because of the relatively large mass of by-products or wastes generated in primary processes, mass allocation has a significantly greater impact than economic allocation. This can certainly discourage producers for implementing such materials in making green concrete. However, since there is no market for CSA in Serbia, it was impossible to perform economic allocation between primary and secondary processes and copper slag was treated as waste in this work. In the basic scenario, the following transport distances were assumed. The concrete is produced in the concrete plant 'Metalka' in Bor, which is located 5 km away from a copper slag plant. Cement is transported by heavy trucks (16-32 t) from the cement factory 'CRH' – Popovac and the estimated distance is 50 km, while river aggregate is transported by medium-sized ships from 'Nova Separacija' – Paracin at an estimated distance of 50 km. Transport distances for each constituent material were doubled to account for the return trip.

3.3 Life cycle impact assessment - LCIA

Impact category indicators related to green-house gases and gases from burning fossil fuels were chosen in this study: global warming potential (GWP), eutrophication potential (EP), acidification potential (AP), and photochemical oxidant creation potential (POCP). They were calculated using the midpoint CML (the Institute of Environmental Sciences of the Faculty of Sciences of Leiden University) baseline methodology [27]. Besides, cumulated energy consumption within the studied part of the life cycle was calculated and expressed as energy use (EU). The LCI and LCIA calculations for each selected mixture were performed using original Excel-based software.

3.4 Sensitivity analysis

A sensitivity analysis was conducted to estimate the impact of the type of used NA and transport distance of CSA. Impacts were calculated for the case where NA was crushed stone instead of river aggregate, assuming the same mix design of concrete. Also, transport distances for CSA were varied from 5 km (as in the basic scenario) to 50 km, which is the assumed transport distance of other concrete constituent materials. Finally, impacts were calculated for the full technological scheme of processing (Figure 1), in which case the electricity consumption is 2.16 kWh (7.78 MJ), while for the internal services, 0.704 l of diesel (24.8 MJ) is used per tone of aggregate. Results are presented in chapter 4.

4 Results and discussion

The impact category indicators calculated for the basic scenario are presented in Figure 4.

The differences between the impacts of NAC and CSAC with both replacement ratios are within a few percent, they are practically equal. This is not surprising since the cement content in all alternatives is the same and cement is by far the largest contributor to all impacts. However, 394 kg/m³ and 529 kg/m³ of NA are preserved in the cases of CSAC_{20_50} and CSAC_{50_50}, respectively. In those cases, the generated waste is reduced by the same amount.

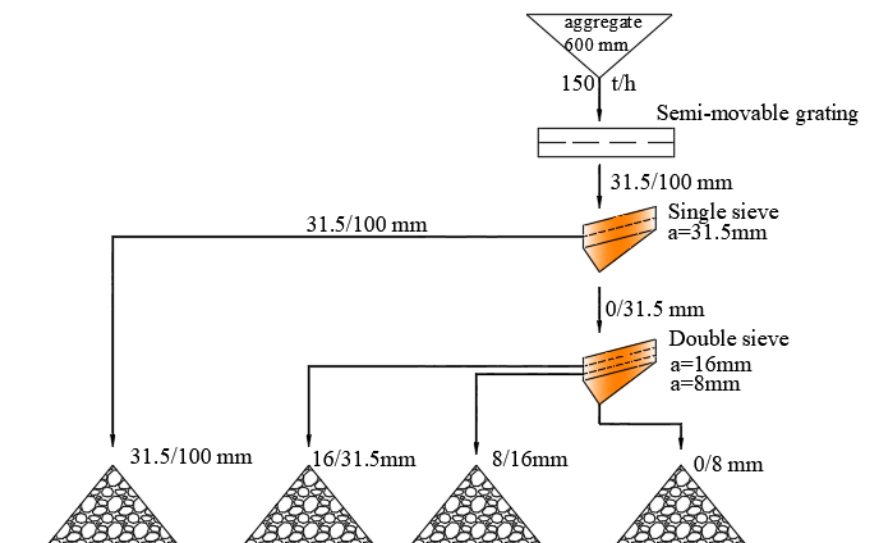


Figure 3. Sieving of copper slag at Mining and Smelting Basin Bor – basic scenario

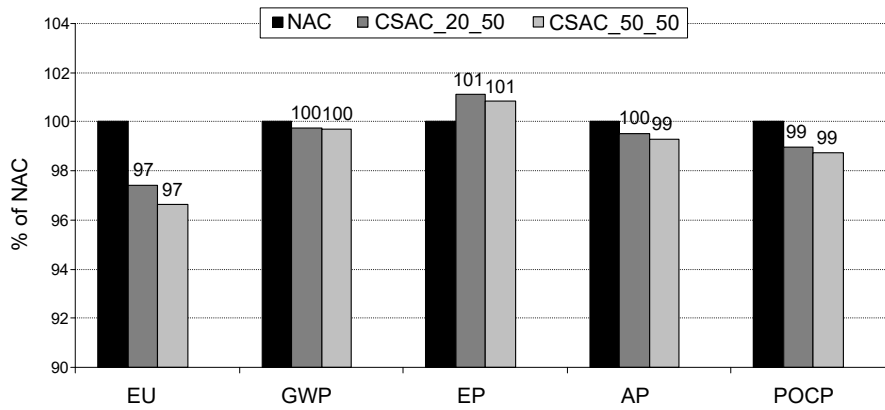


Figure 4. Impact category indicators per FU – basic scenario

If crushed NA is used instead of a river aggregate, the benefit of CSAC in energy use is greater, while other impact categories are similar, Figure 5. This is mostly the consequence of changed transportation type – crushed aggregates are transported by trucks instead of barges as was the case with river aggregate – which affects the NAC impacts more than CSAC impacts.

If the transportation distance of CSA in the basic scenario is changed from 5 km to 50 km, the calculated impact categories are shown in Figure 6. Energy use is now larger in the CSAC case, with all other impacts slightly larger. For all impacts to remain practically equal to the NAC impacts, the transportation distance of CSA should not be greater

than 20 km. Therefore, environmental benefits with CSAC through NA preservation and waste reduction can be obtained if the concrete is produced close to the CSA plant, within a distance of 20 km.

Finally, if the full technological processing scheme (Figure 1) for CSA production is applied, impacts are shown in Figure 7.

Impacts are very similar to those obtained in the basic scenario since the only difference is the larger energy use in the CSA production phase. However, this phase contribution is very small and cannot change total impacts by more than a few percent.

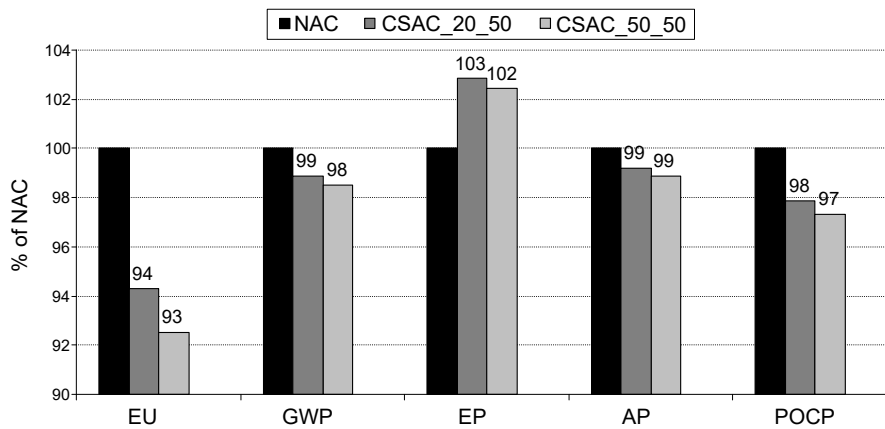


Figure 5. Impact category indicators per FU – crushed NA

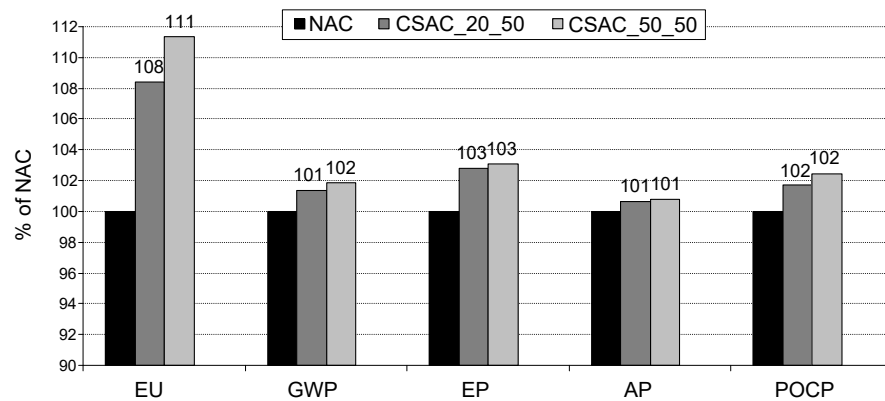


Figure 6. Impact category indicators per FU – all transport distances equal to 50 km

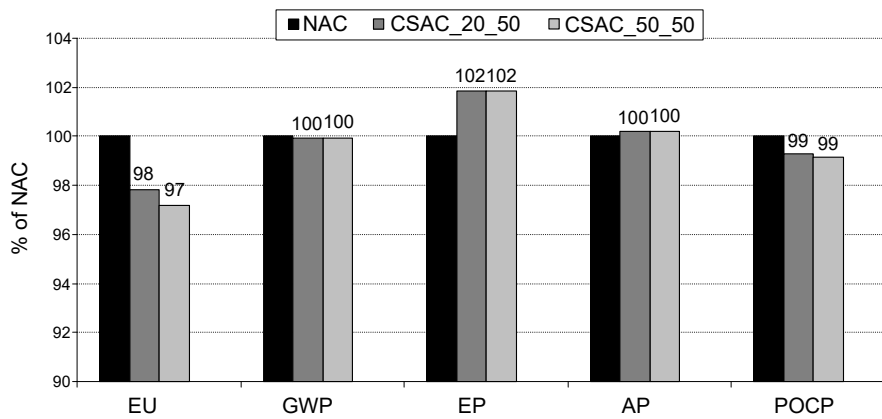


Figure 7. Impact category indicators per FU –full processing scheme

5 Conclusion

In this paper, the environmental potential of replacing the NA with aggregates produced from waste copper slag in concrete mix design was investigated. The conducted case study was based on Serbian LCI data and local conditions in the vicinity of the town of Bor in Serbia. Within these limits, and for chosen impact categories, it was concluded that CSA application in concrete can bring environmental benefits over NA application, in the form of NA preservation and waste reduction. This, however, is valid if the transport distances of CSA are lower than 20 km, so tested concrete mixtures are recommended for local applications.

The amount of waste materials used for the production of CSAC_50_50 makes up about 20% of the concrete unit weight. Yet this concrete can be used in structural applications where low-to-middle strength concrete is economically justified (for instance, in residential buildings). Having this in mind, it can be concluded that local applications of CSAC contribute to the preservation of natural bulk resources and landfill capacity, both becoming an important and scarce resource nowadays in many countries.

Acknowledgment

This work was supported by the Ministry for Education, Science and Technological Development of the Republic of Serbia, research project Grant No. 451-03-9/2021-14/200052 and No. 200092, Faculty of Civil Engineering University of Belgrade. This support is gratefully acknowledged.

References

- [1] C. Meyer, The Greening of the Concrete Industry, *Cement and Concrete Composites* 31 (2009) 601–605.
- [2] Q. Zhao, X. Liu, J. Jiang, Effect of Curing Temperature on Creep Behavior of Fly Ash Concrete, *Construction and Building Materials* 96 (2015) 326–333.
- [3] K.S. Al-Jabri, M. Hisada, A.H. Al-Saidy, S.K. Al-Oraimi, Performance of High Strength Concrete Made with Copper Slag as a Fine Aggregate, *Construction and Building Materials* 23 (2009) 2132–2140.
- [4] J. de Leeuw, D. Shankman, G. Wu, W.F. de Boer, J. Burnham, Q. He, H. Yesou, J. Xiao, Strategic Assessment of the Magnitude and Impacts of Sand Mining in Poyang Lake, China, *Regional Environmental Change* 10 (2010) 95–102.
- [5] G. Kuntikana, D.N. Singh, Contemporary Issues Related to Utilization of Industrial Byproducts, *Advances in Civil Engineering Materials* 6 (2017) 20160050.
- [6] P.S. Prasad, G.V. Ramana, Feasibility Study of Copper Slag as a Structural Fill in Reinforced Soil Structures, *Geotextiles and Geomembranes* 44 (2016) 623–640.
- [7] A. Maharishi, S.P. Singh, L.K. Gupta, Shehnazdeep, Strength and Durability Studies on Slag Cement Concrete Made with Copper Slag as Fine Aggregates, *Materials Today: Proceedings* 38 (2021) 2639–2648.
- [8] D. Urosevic, Copper Extraction from Smelting Slag by Combined Methods, The University of Belgrade, Technical Faculty in Bor, 2016.
- [9] S. Filipović, O. Đokić, A. Radević, D. Zakić, Copper Slag of Pyroxene Composition as a Partial Replacement of Natural Aggregate for Concrete Production, *Minerals* 11(2021) 439.
- [10] B. Gorai, R.K. Jana, Premchand, Characteristics and Utilisation of Copper Slag—a Review. *Resources, Conservation and Recycling* 39 (2003) 299–313.
- [11] C. Shi, C. Meyer, A. Behnood, Utilization of Copper Slag in Cement and Concrete. *Resources, Conservation and Recycling* 52 (2008) 1115–1120.
- [12] R. Neves, F. Branco, J. de Brito, Field Assessment of the Relationship between Natural and Accelerated Concrete Carbonation Resistance, *Cement and Concrete Composites* 41 (2013) 9–15.
- [13] M. Najimi, J. Sobhani, A.R. Pourkhorshidi, Durability of Copper Slag Contained Concrete Exposed to Sulfate Attack. *Construction and Building Materials* 25 (2011) 1895–1905.
- [14] K.S. Al-Jabri, R.A. Taha, A. Al-Hashmi, A.S. Al-Harthy, Effect of Copper Slag and Cement By-Pass Dust Addition on Mechanical Properties of Concrete, *Construction and Building Materials* 20 (2006) 322–331.
- [15] A. Dey, D. Dev, P. Saha, Use of Copper Slag as Sustainable Aggregate, *International Conference on Sustainable Civil Infrastructure* 1 (2014) 229–240.

- [16] K.S. Al-Jabri, M. Hisada, S.K. Al-Oraimi, A.H. Al-Saidy, Copper Slag as Sand Replacement for High Performance Concrete, Cement and Concrete Composites 31 (2009) 483–488.
- [17] A. Rajasekar, K. Arunachalam, M.Kottaisamy, Assessment of Strength and Durability Characteristics of Copper Slag Incorporated Ultra High Strength Concrete, Journal of Cleaner Production 208 (2019) 402–414.
- [18] A. R. Lori, A. Hassani, R. Sedghi, Investigating the mechanical and hydraulic characteristics of pervious concrete containing copper slag as coarse aggregate, Construction and Building Materials, 197 (2019) 130–142.
- [19] Achudhan, Deepavarsa, Vandhana, Khalida, Effect of Copper slag in Structural behaviour of Reinforced Concrete Beams, Materials Today: Proceedings, 5 (2018) 6878–6887.
- [22] I. Ugur, S. Demirdag, H. Yavuz, Effect of Rock Properties on the Los Angeles Abrasion and Impact Test Characteristics of the Aggregates, Materials Characterization 61 (2010) 90–96.
- [21] Q. Yuan, M. Santhanam, Erratum to: Test Methods for Chloride Transport in Concrete, (2013) E1–E1.
- [22] ISO, Environmental Management – Life Cycle Assessment, Set of International standards: ISO 14040-14043, Geneva, International Organization for Standardization, 2006.
- [23] S. Marinkovic, V. Radonjanin, M. Malesev, I. Lukic, Life Cycle Environmental Impact Assessment of Concrete, in L. Bragança, H. Koukkari, R. Blok, H. Gervasio, M. Veljkovic, Z. Plewako, R. Landolfo, V. Ungureanu, L.S. Silva, P. Haller (eds.), Sustainability of Constructions – Integrated Approach to Life-time Structural Engineering. COST action C25. Proceedings of Seminar: Dresden, 6-7 October 2008. Possendorf, Herstellung, Addprint AG, 3.5-3.16.
- [24] M. Spielmann, C. Bauer, R. Dones, M. Tuchschnid, Transport Services. Ecoinvent report no. 14., Swiss Cent. Life Cycle Invent., 2007.
- [25] R. Dones, C. Bauer, R. Bolliger, B. Burger, T. Heck, A. Röder, Paul Scherrer Institut, M.F. Emmenegger, R. Frischknecht, N. Jungbluth, M. Tuchschnid, ESU-services Ltd., Life cycle inventories of energy systems: results for current systems in Switzerland and other UCTE countries, Ecoinvent report. Dübendorf, 2007.
- [26] European Union, Directive 2008/98/EC of the European parliament and of the council on waste and repealing certain directives. Off J Eur Union L312, 2008, 3-30.
- [27] J.B. Guinée, M. Gorée, R. Heijungs, G. Huppes, R. Kleijn, A. de Koning, L. van Oers, A. Wegener Sleswijk, S. Suh, H.A. Udo de Haes, H. de Bruijn, R. van Duin, M.A.J. Huijbregts, 2002. Handbook on life cycle assessment. Operational guide to the ISO standards. I: LCA in perspective. IIa: Guide. IIb: Operational annex. III: Scientific background. Kluwer Academic Publishers, Dordrecht, 2002.