Metallurgical Sludge as Sand Replacement and Constituent of Crushed Concrete Aggregate

Mohamed Alwaeli 1, Jacek Golaszewski 2, Jan Pizon 2, Ada Andrzewska 2, Patryk Szwon 2, Karolina Ceglarz 1, Wojciech Buchta 1, Tomasz Jachnik 1, Jakub Zlotos 1

1 Silesian University of Technology, Faculty of Energy and Environmental Engineering, Konarskiego 18, Gliwice, Poland
2 Silesian University of Technology, Faculty of Civil Engineering, Akademicka 5, Gliwice, Poland

j.pizon@polsl.pl

Abstract. Wastes are indispensable part of human existence. In 2017 in Poland, as much as 90.5% (113.8 million Mg) of all wastes were those related to economic activity. Already for years, the largest group of industrial waste are those from the mining, energy and metallurgical industries. Numerous scientific studies have shown possibilities of using metallurgical waste for the production of concrete and aggregates. What's more, their use can significantly reduce the negative impact on the environment, such as acidification, eutrophication, greenhouse effect or photochemical potential of ozone formation. The aim of the following research is to assess the possibility of using mixture of sludges from the metallurgical industry as a partial replacement for natural aggregates in order to reduce the negative impact on the environment as well as reduce the consumption of natural resources in accordance with the Circular Economy concept. Concrete samples containing 33% of dried and ground sludge as a substitute for sand and concrete containing crushed aggregate from concrete containing this sludge as a substitute for natural coarse aggregate in the amount of 25 and 50% and rounded aggregate from recycled concrete mix containing this sludge as a substitute for natural coarse aggregate in quantities 25 and 50%. The tests of air content, consistency and density of concrete mix as well as compressive strength tests after 2, 7 and 28 days of curing, permeability and water absorption after 28 days of curing were carried out. The sludge as partial sand replacement deteriorates the consistency of concrete mixtures, increases the air content and the density of the concrete mix. Use of recycled concrete aggregates containing sludge also deteriorated the consistency and increased the air content, but at the same time slightly reduced the density of the concrete mix. The use of sludge as a substitute for sand or recycled concrete aggregate as a replacement of natural aggregate affected the absorption of concrete. It is impossible to determine the correlation of absorbability and aggregate replacement level. The use of sludge as a sand replacement has increased the permeability of concrete to the greatest extent. The use of rounded aggregate from recycled concrete mix also but to a lesser extent, and the use of crushed recycled concrete aggregate from did not affect the permeability of concrete. In comparison of compressive strength results of reference concrete, an increase in strength by about 20% for concretes with recycled aggregate and about 40% for concrete with sludge as sand replacement was found. In addition, the conducted research has shown that it is possible to make concrete with the use of metallurgical sludge as a 30% replacement of sand, and then to create aggregate from the resulting composite.
1. Introduction
Wastes are indispensable part of human existence. In 2017 in Poland, as much as 90.5% (113.8 million Mg) of all wastes were those related to economic activity. Already for years, the largest group of industrial waste are those from the mining, energy and metallurgical industries. The aim of the following research is to assess the possibility of using mixture of sludges from the metallurgical industry as a partial replacement for natural aggregates in order to reduce the negative impact on the environment as well as reduce the consumption of natural resources in accordance with the Circular Economy concept.

Numerous scientific studies have shown possibilities of using metallurgical waste for the production of concrete and aggregates. What's more, their use can significantly reduce the negative impact on the environment, such as acidification, eutrophication, greenhouse effect or photochemical potential of ozone formation.

The most widely used metallurgical by-product in concrete technology is blast furnace slag. It is formed during pig iron smelting process. 300 kg of slag is created for every 1000 kg of pig iron [1]. Dependently on cooling method it may take different forms [2]. Lump slag is formed during slow air cooling. It may be used as an aggregate for concrete or in pavement substructures. Expanded slag is formed during cooling under specified thermal and pressure conditions with appropriate amounts of air and water. It is used as lightweight aggregate for concrete. Granulated blast furnace slag is formed during rapid cooling. Ground granulated blast furnace slag (GGBFS) it is commonly used as mineral additive for cement and concrete. It has latent hydraulic properties [3]. Blast furnace slag aggregate (BFSA) may be used for high-strength concrete manufacturing. BFSA concrete have reached 60 – 80% higher compressive strength than traditional concrete, depending on water/cement ratio [4]. Blast furnace slag aggregate may be partial sand replacement for self-compacting concrete [5]. For all replacement levels (10, 25 and 40%), compressive strength was greater in every testing term (7, 28 and 91 days).

Steel slags are created in oxygen converter process. They are richer in SiO₂ and Al₂O₃ and shorter on CaO and Fe₂O₃ in comparison to blast furnace slag and have higher pH value [6]. They may be used as pavements substructures, as a raw material in cement manufacturing or as additive for concrete [7]. Dependently on replacement degree of fine aggregate with low CaO steel slag, the compressive strength of concrete was 110-130% and tensile strength was 140-240% greater in comparison to reference concrete. Optimal replacement was 15-30% for compressive strength and 30-50% for tensile strength [8]. Another researchers have given an information about influence of steel slag aggregate on workability. Consistency was much worse than for reference mix [7]. Optimal replacement level was established as 30-40%.

Steelmaking sludge is formed during wet top blast furnace or converter gas cleaning process. It contains significant amount of iron bearing components, silica and calcium oxides and carbon. If iron content is low it may be used as raw material for cement manufacturing [9]. Essential component of steelmaking sludge is zinc. It leads to cement setting and hardening delay. It reacts with C₃S [10] which results in formation of Ca(Zn(OH))₂ · 2H₂O phase [11]. Lightweight aggregates with 45% of stainless or carbon steel production sludge and 55% of clay was manufactured. Sludge pellets sintered in lower temperature (1050°C) have obtained greater compressive strength than those sintered in higher (1150°C). Better performance was reached by carbon steel sludge pellets than for stainless steel one [12]. Blocks composed of concrete waste, lime production waste and aluminium anodizing sludge (AAW) were subjected to axial strength tests. They were composed of 55-85% of recycled concrete, 10-30% of lime production waste and 10-15% of AAW. Sample with optimal proportions reached greater compressive strength than stated in standard demands [13].
Colangelo et al. [14] have reported influence of natural aggregate for concrete replacement with four types of wastes – construction and demolish wastes, blast furnace slag, marble sludge and fly ash [14]. Influence on environment of such concretes was established. Manufacturing of traditional concrete has the greatest adverse impact on environment. The best solution is to use blast furnace slag in terms of carcinogenic action, air pollution with organic and inorganic compounds, influence on climatic changes, toxicity, eutrophication and natural fuels consumption. Beneficial in terms of air pollutions and ground pollutions are construction and demolish wastes as partial or full replacement of natural aggregates.

Faleschini et al. [15] have performed life cycle analysis (LCA) of electric arc furnace slag (EAFS) as partial replacement of natural aggregates for concrete [15]. They have taken into consideration climatic changes, eutrophication, acidification, oxidants formation, toxicity and influence on ozone layer. They have reported that utilisation of wastes may allow to lower the emissions by about 60% in case of climatic changes, acidification, toxicity and oxidants formation, by 40% in case of eutrophication and toxicity for humans. Utilisation of wastes may allow to lower the ozone layer degradation by about 97%.

Turk et al. [16] have performed life cycle analysis (LCA) of electric arc furnace slag (EAFS), fly ash and moulding sand as constituents of concrete [16]. Usage of those wastes as alternative materials is beneficial for environment because of cement and natural aggregates usage decrease. They have taken into consideration factors of eutrophication, acidification, global warming and ozone formation. Usage of fly ash may reduce concretes impact on environment even by 30% in case of all factors mentioned above. Usage of moulding sand and electric arc furnace slag led to slightly lower results.

2. Materials

Concrete samples containing 33% of dried and ground sludge as a substitute for sand and concrete containing crushed aggregate from concrete containing this sludge as a substitute for natural coarse aggregate in the amount of 25 and 50% and rounded aggregate from recycled concrete mix containing this sludge as a substitute for natural coarse aggregate in quantities 25 and 50% were prepared. Mix of used steelmaking sludge consist of blast furnace, convertor and sintering sludge. Chemical compositions are given in table 1.

|                | Fe  | FeO | SiO₂ | CaO | MgO | Al₂O₃ | Mn   | P₂O₅ | Na₂O | K₂O | Zn | S  | C  | Pb  | ign. loss |
|----------------|-----|-----|------|-----|-----|-------|------|------|------|-----|----|----|----|-----|----------|
| Sintering sludge | 51.6 | 43.0 | 2.1  | 6.6 | 1.3 | 0.9   | 0.5  | 0.09 | 0.13 | 0.13 | 1.8 | 0.3 | 2.3 | 0.2 | 0.9 |
| Blast furnace sludge | 28.5 | 10.3 | 5.2  | 2.5 | 0.6 | 1.4   | 0.2  | 0.09 | 0.12 | 0.18 | 7.3 | 1.3 | 34.5 | 0.6 | 37.7 |
| Converter sludge | 57.7 | 71.4 | 2.4  | 8.3 | 1.2 | 0.6   | 0.8  | 0.12 | 0.21 | 0.13 | 4.9 | 0.2 | 2.8 | 0.3 | 2.1 |

Firstly, four test mortars were prepared in order to determine optimal composition of concrete for crushed and rounded aggregate. Those mortars contained Portland cement, sand, dried sludge as partial sand replacement, water and superplasticizer. Compressive strengths of mortars were tested. Exact composition and compressive strength tests results are given in table 2. Mortar 1 was chosen to prepare final recycled concrete aggregate.

Next, crushed and rounded concrete aggregates were prepared. They consisted of dried sludge, Portland cement, sand, water and superplasticizer. Composition of concrete mixes for aggregate preparation are given in table 3. Rounded recycled concrete aggregate was prepared with usage of RE-CON ZERO admixture and formed in mixer accordingly to manufacturer manual and concrete mix for
crushed aggregate was cast into container where it hardened and broken afterwards. Both rounded and crushed aggregate was cured in water for 28 days. Sieve analysis for those aggregates were conducted as well as for natural aggregate. Grading of aggregates is given in figure 1.

| Table 2. Composition of preliminary recycled concrete aggregates. |
|---------------------------------------------------------------|
|                | Mortar 1 | Mortar 2 | Mortar 3 | Mortar 3 |
| Portland cement CEM I 42,5R [kg/m3] | 450      | 450      | 450      | 450      |
| Water/cement ratio | 0.6      | 0.6      | 0.7      | 0.6      |
| Aggregate (sand + dried sludge) | 1360     | 1360     | 1360     | 1360     |
| Replacement of sand by dried sludge [%] | 30       | 20       | 30       | 30       |
| Superplasticizer [% c.m.] | 1.5      | 1.5      | 1.5      | 3.0      |
| Compressive strength after 28 days [MPa] | 45.0     | 37.7     | 35.2     | 36.0     |

| Table 3. Final composition of recycled concrete aggregates [kg/m³]. |
|------------------------------------------------------------------|
| Portland cement CEM I 42,5R | water | sand | dried sludge | superplasticizer | RE-CON ZERO A component | RE-CON ZERO B component |
| Crushed aggregate | 450 | 270 | 950 | 410 | 13,5 | - |
| Rounded aggregate | 450 | 270 | 950 | 410 | 13,5 | 0,5 | 1,5 |

| Figure 1. Grading curves of natural and recycled concrete aggregates. |

Concrete mixes were prepared with Portland cement CEM I 42,5R, water, sand, dried sludge as a partial replacement of sand, natural aggregate and crushed or rounded recycled concrete aggregate as a partial replacement of natural coarse aggregate. Exact compositions of concrete mixes are given in table 4.
Table 4. Composition of concrete mixes [kg/m^3].

| Concrete Type                  | Symbol | CEM I 42,5R | Water | Sand  | Dried Sludge | Natural Aggregate | Recycled Crushed Aggregate | Recycled Rounded Aggregate |
|-------------------------------|--------|-------------|-------|-------|--------------|--------------------|---------------------------|---------------------------|
| Reference concrete            | REF    | 350         | 210   | 448   | -            | 1342               | -                         | -                         |
| Sludge concrete               | S30    | 350         | 210   | 314   | 134          | 1342               | -                         | -                         |
| Crushed aggregate concrete    | C25    | 350         | 210   | 448   | -            | 1006.5             | 335.5                     | -                         |
| Crushed aggregate concrete    | C50    | 350         | 210   | 448   | -            | 671                | 671                       | -                         |
| Rounded aggregate concrete    | R25    | 350         | 210   | 448   | -            | 1006.5             | -                         | 335.5                     |
| Rounded aggregate concrete    | R50    | 350         | 210   | 448   | -            | 671                | -                         | 671                       |

3. Methods

The tests of air content, consistency and density of concrete mix as well as compressive strength, permeability and water absorption of concrete tests were carried out.

Samples of mortars preliminarily prepared for obtaining optimal recycled aggregate composition were prepared according to standard PN-EN 196-1:2016-07, cured in water of temperature 20±1ºC and tested for compressive strength after 28 days.

Samples of concrete for compressive strength, permeability and water absorption tests were prepared as cubic samples with dimensions 15x15x15 cm. They were cured in water of temperature 20±1ºC and tested for compressive strength after 2, 7 and 28 days and for water absorption and permeability after 28 days.

Tests were conducted according to standards:
- Sieve analysis of aggregates: PN-EN 12620+A1:2010,
- Consistency of concrete mix: PN-EN 12350-2:2011,
- Air content in concrete mix: PN-EN 12350-7:2011,
- Density of concrete mix: PN-EN 12350-6:2011,
- Compressive strength of concrete: PN-EN 12390-3:2011,
- Water absorption of concrete: PN-EN 13369:2018-05,
- Density of concrete: PN-EN 12390-7:2011,
- Permeability of concrete: PN-EN 12390-8:2011.

4. Results and discussion

4.1. Concrete mix properties

Results of slump, air content and density tests of concrete mix are given in table 5. Reference concrete mix has achieved 10.5 cm of slump, which corresponds with S3 consistency class. Modification of concrete mix with dried sludge or recycled concrete aggregate has caused decrease of consistency. Mix with dried sludge as partial replacement of sand achieved 3 cm of slump which classifies it as S2 consistency class. Mixes containing 25% of crushed or round recycled concrete aggregates were of S2 class with slump 6 and 5 cm respectively. Mixes containing 25% of crushed or round recycled concrete aggregates have achieved 0 cm slump and they have not meet standard demands for any
consistency class. It means that without plasticizing agents it is impossible to make workable concrete mix.

Density of concrete mix containing ground dried sludge as partial replacement of sand is slightly greater than of reference concrete mix. It is associated with incorporation of very fine fraction into concrete mix that causes decrease of void ratio in aggregate. Simultaneously, air content in concrete mix containing dried sludge is slightly higher. It is connected with consistency decrease and difficulties with compaction and deaerating of concrete mix. Density drops and air content increase with raising amount of crushed or rounded recycled concrete aggregate as partial replacement of natural coarse aggregate in concrete. It is connected with consistency decrease and difficulties with proper compaction of concrete mix.

### Table 5. Concrete mixes properties

| Symbol | Slump [cm] | Air content [%] | Density [kg/m³] |
|--------|------------|-----------------|-----------------|
| REF    | 10.5       | 1.5             | 2.32            |
| S30    | 3.0        | 2.5             | 2.40            |
| C25    | 6.0        | 2.9             | 2.25            |
| C50    | 0.0        | 3.0             | 2.27            |
| R25    | 5.0        | 2.9             | 2.29            |
| R50    | 0.0        | 4.0             | 2.25            |

### 4.2. Compressive strength

Compressive strength of concretes tests results are given in figures 2-4. After 2 days of curing reference concrete has achieved 12.6 MPa. Compressive strength of concrete containing dried sludge as partial replacement of sand was 17.1 MPa. Better effects were obtained in case of partial replacement of coarse aggregate with recycled concrete aggregate. Concrete containing crushed recycled concrete aggregate have achieved 25.6 and 30.4 MPa for replacement level of 25 and 50% respectively. Those containing rounded recycled concrete mix aggregate have achieved 17.5 and 24.5 MPa for replacement level of 25 and 50% respectively. Compressive strength increase in comparison to reference concrete is connected with lowered effective water/cement ratio due to higher water absorbability of recycled concrete aggregates in comparison to natural one. Concretes with crushed recycled concrete aggregate gained more compressive strength than those with rounded recycled concrete mix aggregate.

![Figure 2. Compressive strength of concrete after 2 days.](image-url)
After 7 days of curing reference concrete has gained 28.2 MPa. Compressive strength of concrete containing dried sludge as partial replacement of sand was 40.0 MPa. Concrete containing crushed recycled concrete aggregate have achieved 43.7 and 45.3 MPa for replacement level of 25 and 50% respectively. Those containing rounded recycled concrete mix aggregate have achieved 38.2 and 41.2 MPa for replacement level of 25 and 50% respectively. Effects obtained in cases of partial substitution of sand with dried sludge and of partial replacement of coarse aggregate with recycled concrete aggregate were similar. Compressive strength increase in comparison to reference concrete is connected with lowered effective water/cement ratio. Concretes with crushed recycled concrete aggregate gained more compressive strength than those with rounded recycled concrete mix aggregate. It is connected with grain shape and possibly with granulating agent presence.

After 28 days of curing reference concrete has gained 40.6 MPa. Compressive strength of concrete containing dried sludge as partial replacement of sand was 56.3 MPa. Concrete containing crushed recycled concrete aggregate have achieved 49.7 and 49.4 MPa for replacement level of 25 and 50% respectively. Those containing rounded recycled concrete mix aggregate have achieved 50.0 and 48.4 MPa for replacement level of 25 and 50% respectively. Better effects were obtained in case of partial replacement of sand with dried sludge than in case of partial replacement of coarse aggregate with recycled concrete aggregate. Additional compressive strength increase of concrete with recycled concrete aggregate in comparison to reference concrete may be also connected with additional curing of aggregate inside the concrete. In real case however it will not take place because concrete structures to be demolished and recycled are almost always fully cured.
4.3. Other hardened concrete mix properties

Results from other hardened concrete tests such as permeability, water absorption and density are given in table 6. Density of hardened concrete containing ground dried sludge as partial replacement of sand is similar to reference concrete. Density slightly decreases with raising amount of crushed or rounded recycled concrete aggregate as partial replacement of natural coarse aggregate in concrete. It is connected with lower density of recycled concrete aggregate itself in comparison to natural one.

Incorporation of dried sludge as partial replacement of sand in concrete has not influenced water absorbability of concrete. Usage of recycled concrete aggregates however has raised water absorbability. It is associated with higher water absorbability of recycled concrete in comparison to natural aggregates and higher air content in modified concretes.

Permeability of concrete containing crushed recycled concrete aggregate as 25% replacement of natural coarse aggregate is similar to reference concrete. Raising of crushed recycled concrete aggregate content leads to slight increase of permeability. Permeability of concrete containing rounded recycled concrete mix aggregate as 25% replacement of natural coarse aggregate is also similar to reference concrete. Raising of rounded recycled concrete aggregate content leads to significant increase of permeability.

| Symbol | Permeability [mm] | Water absorption [%] | Density [kg/m³] |
|--------|-------------------|----------------------|-----------------|
| REF    | 3                 | 6.54                 | 2.19            |
| S30    | 19                | 6.50                 | 2.21            |
| C25    | 1                 | 7.12                 | 2.16            |
| C50    | 7                 | 4.93                 | 2.10            |
| R25    | 2                 | 6.39                 | 2.16            |
| R50    | 80                | 7.86                 | 2.15            |

5. Conclusions

The sludge as partial sand replacement deteriorates the consistency of concrete mixtures, increases the air content and the density of the concrete mix. Use of recycled concrete aggregates containing sludge also deteriorated the consistency and increased the air content, but at the same time slightly reduced the density of the concrete mix.

The use of sludge as a substitute for sand or recycled concrete aggregate as a replacement of natural aggregate affected the absorption of concrete. It is impossible to determine the correlation of absorbability and aggregate replacement level.

The use of sludge as a sand replacement has increased the permeability of concrete to the greatest extent. The use of rounded aggregate from recycled concrete mix also but to a lesser extent, and the use of crushed recycled concrete aggregate from did not affect the permeability of concrete.

In comparison of compressive strength results of reference concrete, an increase in strength by about 20% for concretes with recycled aggregate and about 40% for concrete with sludge as sand replacement was found. In addition, the conducted research has shown that it is possible to make concrete with the use of metallurgical sludge as a 30% replacement of sand, and then to create aggregate from the resulting composite. Thus such actions dealing with partial or full replacement of natural aggregates for concrete form a part of the Circular Economy concept.
Acknowledgments
This work received support from the PROM Programme - International scholarship exchange of PhD students and academics financed by National Agency for Academic Exchange and the European Social Fund under the Operational Program Knowledge Education Development.

The research reported in this paper was co-financed by the European Union from the European Social Fund in the framework of the project "Silesian University of Technology as a Center of Modern Education based on research and innovation

References
[1] Z. Pater, *Podstawy metalurgii i-odlewnictwa*. Lublin: Politechnika Lubelska, 2014.
[2] W. Chesner, R. Collins, and M. MacKay, “User Guidelines for Waste and Byproduct Materials in Pavement Construction - FHWA-RD-97-148,” 2016.
[3] A. M. Neville, *Properties of Concrete*. Harlow: Pearson Education Limited, 1995.
[4] R. Demirboğa and R. Gül, “Production of high strength concrete by use of industrial by-products,” *Build. Environ.*, vol. 41, no. 8, pp. 1124–1127, Aug. 2006.
[5] G. Singh and R. Siddique, “Strength properties and micro-structural analysis of self-compacting concrete made with iron slag as partial replacement of fine aggregates,” *Constr. Build. Mater.*, vol. 127, pp. 144–152, Nov. 2016.
[6] I. Jonczy and L. Lesia, “Charakterystyka składu chemicznego żużli konwertorowych i wielkopiecowych,” *Górnictwo i Geol.*, vol. 8, no. 4, pp. 51–61, 2013.
[7] V. S. Devi and B. K. Gnanavel, “Properties of Concrete Manufactured Using Steel Slag,” *Procedia Eng.*, vol. 97, pp. 95–104, Jan. 2014.
[8] H. Qasrawi, F. Shalabi, and I. Asi, “Use of low CaO unprocessed steel slag in concrete as fine aggregate,” *Constr. Build. Mater.*, vol. 23, no. 2, pp. 1118–1125, Feb. 2009.
[9] J.-W. Ahn, “Development of Technology for Raw Material of Ordinary Portland Cement and Practical Use of Sludge made from Steel Industry by Complex Treatment,” *Resour. Process.*, vol. 51, no. 1, pp. 42–47, 2004.
[10] M. Gawlicki and D. Czamarska, “Effect of ZnO on the hydration of Portland cement,” *J. Therm. Anal.*, vol. 38, no. 9, pp. 2157–2161, Sep. 1992.
[11] M. A. Trezza, “Hydration study of ordinary portland cement in the presence of zinc ions,” *Mater. Res.*, vol. 10, no. 4, pp. 331–334, Dec. 2007.
[12] Y.-L. Wei and G.-W. Ko, “Use of low CaO unprocessed steel slag in concrete as fine aggregate,” *Constr. Build. Mater.*, vol. 23, no. 2, pp. 1118–1125, Feb. 2009.
[13] J.-W. Ahn, “Development of Technology for Raw Material of Ordinary Portland Cement and Practical Use of Sludge made from Steel Industry by Complex Treatment,” *Resour. Process.*, vol. 51, no. 1, pp. 42–47, 2004.
[14] F. Colangelo et al., “Recycled concrete containing EAF slag: environmental assessment through LCA,” *Waste Manag.*, vol. 45, pp. 194–205, Nov. 2015.