19/2017
Lederer, J.; Bogush, A.; Fellner, J. (2017) „The utilization of solid residues from municipal solid waste incineration in the cement industry: A review” In: CD-Proceedings Sardinia 2017, 16th International Waste Management and Landfill Symposium “Sardinia Symposium 30th Anniversary Book”, 2-6 Oct. 2017, S. Margherita di Pula (Cagliari), Sardinia, Ed.: Cossu, R.; Stegmann, R., CISA Publisher, Sardina, paper 171, p 1-7, ISSN 2282-0027, ISBN 978-88-6265-010-6

THE UTILIZATION OF SOLID RESIDUES FROM MUNICIPAL SOLID WASTE INCINERATION IN THE CEMENT INDUSTRY: A REVIEW

J. LEDERER*, A. BOGUSH**, J. FELLNER*

**Institute for Water Quality, Resource and Waste Management, TU Wien, Karlsplatz 13/226, 1040 Vienna, Austria*
*** Department of Civil, Environmental & Geomatic Engineering, University College London, Chadwick Building, Gower Street, London, WC1E 6BT*

SUMMARY: A number of studies have been carried out on the utilization of residues from municipal solid waste incineration air pollution control (APC), hereafter termed as MSWI fly ashes, in the cement industry, in order to substitute raw materials in cement production. This article provides a review on the current state of knowledge on this type of utilization of secondary raw materials. Results show that studies investigate different utilizations of the residues in cement production, either as secondary raw material for Portland cement clinker production, or as secondary raw material that serves as major or minor cement constituent which can be added to Portland cement clinker (cement blend). Studies also investigate the impact of pre-treatment of residues prior their utilization, particularly washing in order to remove salts. Beside the impact of the addition of the residues to the clinker or cement on the cement quality, environmental considerations are taken into account by studies. However, these are usually limited to leaching tests for heavy metals. Heavy metal and organic pollutants emissions to the air or the accumulation of heavy metals in the product are not taken into account. Future research should also focus on these aspects of the utilization of municipal solid waste incineration in the cement industry.

1. INTRODUCTION

The disposal of residues from municipal solid waste incineration (MSWI), i.e. fly ashes and Air-Pollution-Controll (APC) residues (hereafter termed as MSWI fly ashes) is becoming a growing environmental issue. Common practices like landfilling to hazardous waste landfills or landfilling of pretreated residues to non-hazardous landfills are challenged by concerns on ecological sustainability (i.e. long-term emissions of salts and heavy metals from disposal sites, utilization of considerable amounts of cement for stabilization), financial restraints (i.e. high costs for MSWI fly ash treatment and disposal), and low recycling rates of secondary raw materials from MSWI fly ash (i.e. metals, minerals). In order to deal with these concerns, research ambitions aimed to find alternative ways of MSWI fly ash treatment and particularly utilization as secondary raw material. One of these ambitions focuses on the utilization of MSWI fly ashes rich in CaO, SiO_2, Al_2O_3, and Fe_2O_3 in cement production.

A number of studies have been carried out on the utilization of residues from residues from

Proceedings Sardinia 2017 / Sixteenth International Waste Management and Landfill Symposium/ 2 - 6 October 2017
S. Margherita di Pula, Cagliari, Italy /

municipal solid waste incineration in the cement industry, in order to substitute Portland cement clinker raw materials or cement raw materials in cement blends. This article provides a review on the current state of knowledge on this type of utilization of secondary raw materials.

2. METHODS AND MATERIALS

Scientific databases (i.e. Scopus.com) are used, using the search terms “waste” AND “incineration” AND “ash” AND “cement”. After ruling out the publications not to be considered (e.g. stabilization of waste incineration ashes with cement for landfilling), the remainder is analysed, based on technological, product and environmental parameters. Only a selected number is presented in this article.

3. RESULTS AND DISCUSSION

3.1 Number of articles

Results show that the number of articles has vastly increased during the last years, particularly since the year 2000 (Figure 1). Having a look at the home countries of the authors’ affiliated institutions, China is dominating by far, followed by European countries, Taiwan, and the US. Within Europe, West- and South-European countries are dominating, while Nordic countries or German-speaking countries are not represented at all, indicating a low interest in this topic (Figure 2). This might be due to the legal constraints in these countries. In Switzerland and lately also in Austria, MSWI fly ashes will not or only hardly meet the limit values required for their utilization in cement production (Lederer et al., 2017).

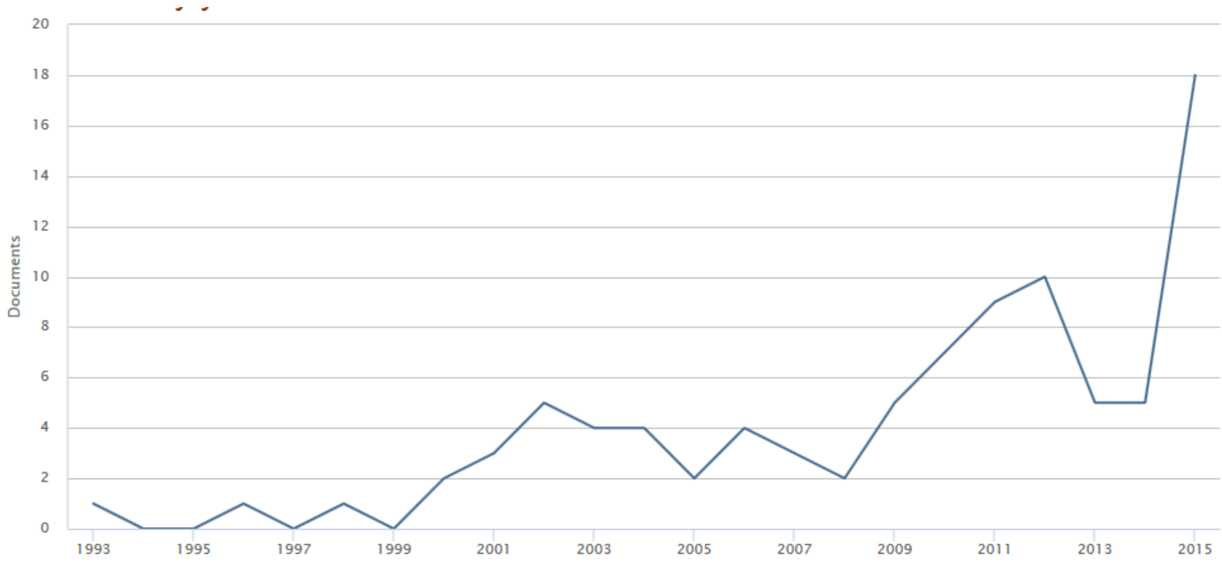

Figure 1. Number of publications on the utilization of MSWI ashes in the cement industry, retrieved from Scopus.com.

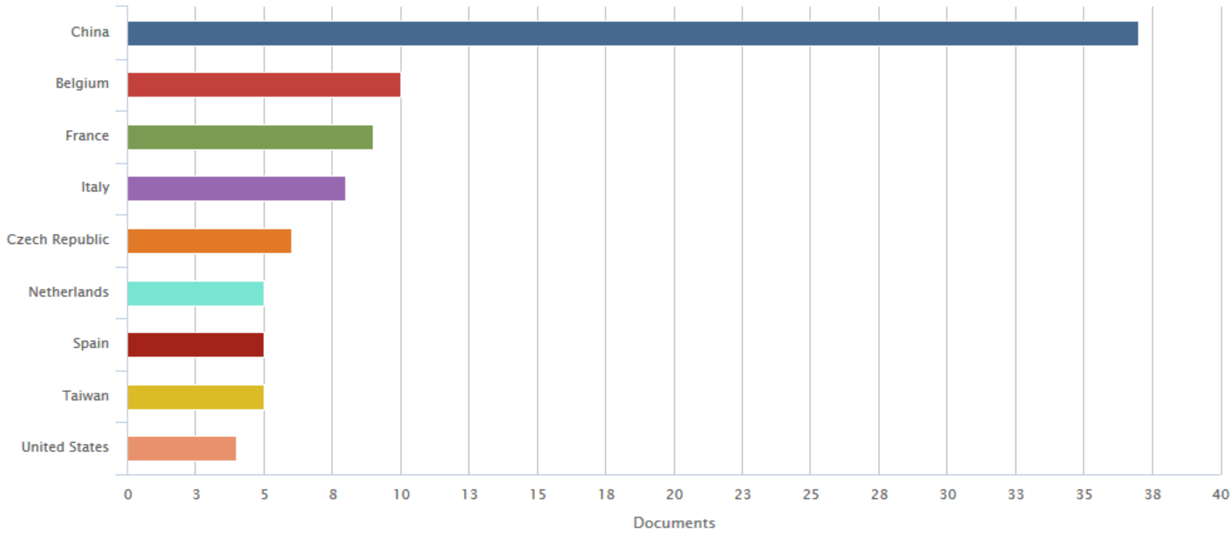

Figure 2. Number of publications on the utilization of MSWI ashes in the cement industry, by country, retrieved from Scopus.com.

3.2 General approaches of utilizing MSWI fly ashes in the cement production

Results show that there are generally four ways of applying of waste incineration residues in the cement industry:

3.2.1 Introduction of untreated MSWI residues in the clinker production

Several studies investigate and suggest the introduction of untreated MSWI fly ash as a secondary raw material to produce clinker in the cement kiln.

Saikia et al. (2007) used untreated and treated (washed) MSWI fly ash collected from the flue gas cleaning to produce a cement clinker at laboratory scale. The treated MSWI fly ashes were washed with a liquid/solid (L/S) ratio of 20. The share of MSWI fly ash was 44-50% of the clinker raw materials. Clinker was produced under 1,300 and 1,400 °C. With regards to heavy metal flows, the authors show that under these temperatures, metals like Cd, Hg or Pb volatilize and find their way to the flue-gas rather than ending up in the clinker. However, in practice, they will evaporate on dust particles which will be partly removed from the flue-gas by the air pollution control (APC) of the cement plant. As the dust from the APC is widely used as secondary milling material, the heavy metals removed from the off-gas earlier will be incorporated in the cement again (Lederer et al., 2017). In their conclusion, the authors argue that it is possible to produce cement clinkers without any pretreatment such as washing, even though the washing reduces chlorine related problems in the operation of the kilns.

Wu et al. (2011) used untreated MSWI fly ashes from the APC in concentrations of 15-44% as clinker raw material to produce clinker (under temperatures of 1,150-1,300 °C) for sulfoaluminate cement production on a laboratory scale. Based on their results, the authors concluded that a maximum share of MSWI fly ash addition of 30% is possible to produce clinker under a temperature of 1,250 °C for two hours. Though the authors also find a strong immobilization of heavy metals in the clinker (based on leaching tests with mortars produced from the clinker), they emphasize further research problems, like the impact of Cl on the corrosion of the cement kiln.

Guo et al. (2015) used 30% of untreated MSWI fly ash to burn clinker under 1,200 °C for alinate cement at laboratory scale. Based on their investigation results on the strength, durability (shrinking, carbonation, water permeation, reinforcement steel corrosion), and environmental risk, the authors conclude that a cement with lower production CO_2 emissions and energy demand can be produced.

What all of these studies have in common is that they positively assess the utilization of untreated and raw MSWI fly ash as secondary raw material to be used for clinker production.

3.2.2 Introduction of washed MSWI fly ashes in the clinker production

Even though suggesting that a utilization of untreated MSWI fly ash as secondary raw material for clinker production in the cement kiln is possible, some of the first set of studies reviewed underline the problems for cement quality and particularly for the cement plant components through the introduction of untreated, high chlorine and sulfate containing MSWI fly ashes in the cement kiln. A possible solution for this problem would be a pre-treatment of the MSWI fly ashes by washing them with water.

This procedure was applied in the aforementioned study of Saikia, et al. (2007), but also by a Taiwanese study by Pan et al. (2008). In their study, MSWI fly and bottom ashes were either washed with water (and a not specified, thus assumed to be negligible amount of acid) in order to reduce the chlorides content. The MSWI fly ashes were added at a concentration of 1.75% of all clinker raw materials, which corresponds to the ratio of MSWI fly ash and cement production in Taiwan. As the addition of the ashes did not affect the cement quality, the authors conclude that by applying the pre-treatment procedure suggested, “MSWI fly ashes […] are suitable for reuse in cement production” (Pan, 2008).

A common finding by all the authors cited is that MSWI fly ashes can be a substitute of clinker raw materials, particularly if chlorides are removed prior the application.

3.2.3 Introduction of untreated MSWI fly ashes to replace cement

Like other residues from thermal processes (e.g. coal fly ash), MSWI fly ashes have properties positively influencing the hydration behavior of certain cements if added to the clinker as secondary cement raw material in the cement mill.

Rémond et al. (2002) investigated the properties of mortars containing up to 20% of untreated and treated (washed) MSWI fly ashes in the underlying cement. Even though an increased MSWI fly ash content lead to higher setting times, the highest long term compressive strength was found for a MSWI fly ash content of 10%. Only at 15% the compressive strength tends to decrease compared to the ordinary control standard cement.

Shi et al. (2009) investigate the heavy metals leaching of mortars produced from cements with 40% untreated MSWI fly ash and 60% Portland cement, finding that the leaching rates of both, surface leaching and crushed mortar leaching, are within acceptable limit range.

Keppert et al. (2012) analyze the physical and chemical properties (composition, morphology, hydration behavior) of different MSWI residues (boiler fly ash, electrostatic precipitator ash (ESP), bottom ash) in comparison to fly ash from a coal fired power plant in order to assess whether they can be utilized for cement production. Based on their results, the authors conclude that the MSWI fly ashes can be used for cement production, but in untreated and raw form, as neither washing for salt removal (which causes loss of the pozzolanic activity), nor thermal processes like vitrification (due to high financial costs) seem to be feasible.

Kim et al. (2015) used untreated fly and bottom ash to replace 10%, 20%, and 30% of cement respectively. Even though all of the MSWI fly ash amended mortar samples reached the limit values for compressive strength, which was found to be even similar to higher than the control mortar, the authors conclude that some pre-treatment would be required. However, this counts more for the bottom than for the fly ashes.

Another study from Taiwan by Huang and Chuieh (2015) performed a Life Cycle Assessment (LCA) for different treatment, disposal, and recycling options for MSWI fly ashes (solidification+landfilling, clinker raw material substitute, brick-making raw material substitute, and Zn recycling). For the utilization of MSWI fly ashes to substitute clinker raw materials, MSW fly ash is washed and added at 10% of clinker input raw materials. Based on their evaluation, this option turned out to be second best out of four, making it a viable option for the authors.

3.2.4 Introduction of washed MSWI fly ashes to replace cement

Unlike for clinker raw materials, the content of water-soluble chlorides in cement raw material is seen critical not because of plant-based corrosion, but due to corrosion of reinforcement steel. Thus, European Standards limit the concentration of chlorides in cement raw materials like coal fly ash to 0.1% (DIN, 2012). Subsequently, and in order to comply with European Standards (even though they don't consider MSWI fly ashes), some chlorides removal prior application is required, most likely through washing the MSWI fly ashes. After drying the washed fly ashes, they can be used as cement substitute.

Rémond et al. (2002) also investigated the properties of mortars containing cements containing 10% of washed, thus chloride and sulfate freed MSWI fly ash. However, the compactive strength was significantly lower than with unwashed MSWI fly ash addition.

Gao et al. (2008) tested different cement mortars with a cement substitution by washed MSWI fly ash (liquid to solid ratio L/S=5, residence time T=30 min) of 10, 20, and 30%, finding the acceptable substitution rate at 20% in order to maintain the strength of the concrete. However, the authors judge the chloride content of 1.3% after washing (reduced from 10% before washing) as too high in order to prevent reinforcement steel corrosion. If the washing process can be improved (e.g. increase of L/S ratio or T), the MSWI fly ashes can be a useful cement substitute.

Hartmann et al. (2015) used raw and washed MSWI fly ashes (L/S=10, T=24 h) to replace cement. The washing showed the following removal rates of Cl: 82% after 1 h, 98% after 2 h, and 99.6% after 24 h. The mortars produced substituted 30% and 50% of cement by washed MSWI fly ashes. Another control mortar substituted 30% of cement by raw fly ashes. Results showed comparable heavy metal leaching values of the 30% amended and the control mortar. Of the MSWI fly ash (raw and washed) amended mortars, only the 30% amended mortar reached the compressive strength limit. For these two reasons, the authors conclude that the washed MSWI fly ash can be a useful substitute of cement raw materials.

3.3 Comparison of approaches

Option 1 has the disadvantage of the release of chloride to the flue gas may lead to corrosion problems in the cement kiln. This is also the reason why cement plant operators prefer refuse derived fuels containg low PVC contents. Otherwise, an energy-intensive chlorine bypass must be introduce in the APC of the cement kiln. Furthermore, heavy metal emissions from the cement kiln might be problematic if no appropriate accompany measures (i.e. APC system and management of APC residues in the cement kiln) are introduced. Therefore, this option has the advantage of making use of a material that does not release additional CO_2, as the $CaCO_3$ or $Ca(OH)_2$ presented or added in the waste or added to the APC system has already been calcined. With regards to the quality of the cement, free lime (CaO) causing lime swelling should not be a big problem, as CaO should be integrated in the cement phases. Na and K contents causing alkali–silica reaction (ASR) in the concrete, however, might be a long term concrete quality problem and require further investigation.

Option 2 reduces the corrosion risk in the cement kiln stack and thus the necessity of chlorine bypass. Also, a potential ASR risk should be reduced, as large parts of Na and K can be removed during the washing. However, beside havin the same problem of heavy metal release, the release of CO_2 might be higher than in Option 1 in case that the CaO present in the MSWI fly ash has already been converted into $CaCO_3$ due to storage after washing. If storage times are not too long, however, it is more likely that the CaO has converted into $Ca(OH)_2$, causing no additional CO_2 emissions if compared to haing solely CaO. The only additional climate impact if compared to CaO comes from the energy used for dehydrating the $Ca(OH)_2$. Another obstacle is the cleaning of the waste water origin from washing, as in practice, large amounts of water might be used.

For Option 3, the gaseous emissions of heavy metals will not be a problem, except for Hg during cement milling. However, the presence of CaO, chlorides, and other concrete damaging compounds will limit the utilization.

Option 4 does not have these high problems with chlorides and other concrete damaging substances, but therefore, also properties beneficial for the concrete hardening are lost due to the washing process.

What all of these options have in common is that depending on the amount of MSWI fly ashes added, the heavy metal contents in concrete will increase. While leaching tests carried out by researchers show that at least in the short run, most heavy metals are fixed in the concrete matrix, acceptable limits for the total contents of heavy metals in cement have to be defined. However, none of the articles defined these acceptable limits based on total contents, as it is the legal rule in countries like Austria and Switzerland (Lederer et al., 2017). Furthermore, none of the articles reviewed considered the gaseous emissions, neither of organic compounds, nor of heavy metals present in MSWI fly ash. This counts particularly for Options 1 and 2 in the case of Cd and Hg, both released during clinker burning (and probably also in the raw mill if added there – however, then, organic pollutants might be a problem too), and Options 3 and 4 for Hg and organic pollutants (released in the cement mill). These two aspects, as well as the long-term product quality with respect to free lime and sulphur compounds, have to be further investigated in future.

CONCLUSIONS

The result of the review suggest that, even though a large number of studies on the utilization of MSWI residues in the cement industry have been carried out, there are still aspects that have not been considered. This not only counts for long-term behaviour and product quality, but also environmental emissions and stock building. Furthermore, more focus should be given at bottom ashes, for which even not enough articles have been found in order to justify their inclusion in this review at hand.

AKNOWLEDGEMENTS

The presented work is part of a large-scale research initiative on anthropogenic resources

(Christian Doppler Laboratory for Anthropogenic Resources). The financial support of this research initiative by the Federal Ministry of Economy, Family and Youth and the National Foundation for Research, Technology and Development is gratefully acknowledged. Industry partners cofinancing the research center on anthropogenic resources are Altstoff Recycling Austria AG (ARA), Borealis Group, Voestalpine AG, Wien Energie GmbH, Wiener Kommunal-Umweltschutzprojektgesellschaft GmbH, and Wiener Linien GmbH & Co KG.

REFERENCES

DIN, 2012. Flugasche für Beton –Teil 1: Definition, Anforderungen und Konformitätskriterien; Deutsche Fassung EN 450-1:2012 (Fly ash for concrete – Part 1: Definition, specifications and conformity criteria; German version EN 450-1:2012), in: Deutsches Institut für Normung (DIN), Berlin.

Gao, X., Wang, W., Ye, T., Wang, F., Lan, Y., 2008. Utilization of washed MSWI fly ash as partial cement substitute with the addition of dithiocarbamic chelate. Journal of Environmental Management 88, 293-299.

Guo, X., Shi, H., Wu, K., Ju, Z., Dick, W., 2015. Performance and risk assessment of alinite cement-based materials from municipal solid waste incineration fly ash (MSWIFA). Mater Struct, 1-9.

Hartmann, S., Koval', L., Škrobánková, H., Matýsek, D., Winter, F., Purgar, A., 2015. Possibilities of municipal solid waste incinerator fly ash utilisation. Waste Management & Research 33, 740-747.

Huang, T.Y., Chuieh, P.T., 2015. Life Cycle Assessment of Reusing Fly Ash from Municipal Solid Waste Incineration. Procedia Engineering 118, 984-991.

Keppert, M., Pavlík, Z., Tydlitát, V., Volfová, P., Švarcová, S., Šyc, M., Černý, R., 2012. Properties of municipal solid waste incineration ashes with respect to their separation temperature. Waste Management & Research 30, 1041-1048.

Kim, J., An, J., Nam, B.H., Tasneem, K.M., 2015. Investigation on the side effects of municipal solid waste incineration ashes when used as mineral addition in cement-based material. Road Materials and Pavement Design, 1-20.

Lederer, J., Trinkel, V., & Fellner, J. (2017). Wide-scale utilization of MSWI fly ashes in cement production and its impact on average heavy metal contents in cements: The case of Austria. Waste Management, 60, 247-258.

Pan, J.R., Huang, C., Kuo, J.-J., Lin, S.-H., 2008. Recycling MSWI bottom and fly ash as raw materials for Portland cement. Waste Management 28, 1113-1118.

Rémond, S., Pimienta, P., Bentz, D.P., 2002. Effects of the incorporation of Municipal Solid Waste Incineration fly ash in cement pastes and mortars: I. Experimental study. Cement Concrete Res 32, 303-311.

Saikia, N., Kato, S., Kojima, T., 2007. Production of cement clinkers from municipal solid waste incineration (MSWI) fly ash. Waste Management 27, 1178-1189.

Shi, H.-S., Kan, L.-L., 2009. Leaching behavior of heavy metals from municipal solid wastes incineration (MSWI) fly ash used in concrete. Journal of Hazardous Materials 164, 750-754.

Wu, K., Shi, H., Guo, X., 2011. Utilization of municipal solid waste incineration fly ash for sulfoaluminate cement clinker production. Waste Management 31, 2001-2008.