Utilization Status of Oil-Based Drilling Cutting Pyrolysis Residues in The Field of Building Materials

Hong Li [1,*], Qionghua Jiang [2], Juan He [3]

[1] Luzhou Vocational ＆ Technical College, Luzhou 646000, China; 505366981@qq.com

[2] Pengshui Tujia and Miao Autonomous County Environmental Monitoring Station, Chongqing 409600, China

[3] Chongqing Jingchuang United Environmental Engineering Co., Ltd., Chongqing 404100, China

* Author to whom correspondence should be addressed.

Abstract. The exploitation of shale gas generates a large amount of oil-based drill cuttings, which are harmful to the environment due to their content of petroleum hydrocarbons, heavy metals, and organic pollutants. Thermal desorption technology is one of the main techniques for treating oil-based drill cuttings. This paper focuses on oil-based drilling cutting pyrolysis residues and summarizes its current research status in the construction materials field. Currently, the application of oil-based drilling cutting pyrolysis residues has achieved certain results in road fill, cement production, ceramsite production, and wall materials, and further exploration and development are needed in the future to develop high value-added products and expand the pathways for the high-value utilization of oil residues.

Keywords: oil-based drilling cutting pyrolysis residues; road fill; cement production; wall materials.

1. Introduction

Shale gas is an unconventional natural gas resource stored in shale formations that can be extracted, with advantages such as long extraction life and production cycle. It is estimated that China's technically recoverable shale gas resources are about 36.1 trillion cubic meters, making it an important energy resource and raw material for the chemical industry. In order to ensure national energy security and optimize the energy structure, the country continues to increase efforts in shale gas development. According to the "Shale Gas Development Plan (2016-2020)", China's shale gas production is expected to reach (800-1000)×10^8 cubic meters per year by 2030. The generated oil-based drilling cuttings will reach (800-1,000)×10^4 tons.

The main contaminants in oil-based drilling cuttings include oil, heavy metals, inorganic salts, and high molecular weight organic compounds, which belong to the hazardous waste category HW08 in the "National Hazardous Waste List" (2021 edition). The thermal desorption technology for oil-based drilling cuttings is widely used in their treatment due to its unique separation and recovery advantages. The oil-based drilling cutting pyrolysis residues (ODCPRs) is mainly used in the construction materials field for road fill, cement production, ceramsite, wall materials, and other applications.

2. Used as road fill

Research on the use of ODCPRs as road fill mainly includes two aspects: as subgrade fill and pavement fill. The main performance indicators for subgrade fill are mechanical properties, therefore, it is necessary to select suitable solidifying agents to solidify ODCPRs and improve the mechanical properties of the fill material. Jiao et al.(2022) used hydrated lime and fly ash as admixtures, replaced some of the natural soil with ODCPRs, and prepared modified subgrade fill materials with different mix ratios. The unconfined compressive strength and California bearing ratio of the fill material can meet the requirements for subgrade and pavement fill strength of high-grade highways and first-grade highways. Ren et al.(2018) used cement as a solidifying agent, and added lime and ODCPRs to prepare subgrade materials. The strength indicators can meet the requirements for paving third and fourth-grade highways or roads around drilling wells, as well as paving well sites, and the leaching of heavy metals, benzo(a)pyrene, petroleum products, COD, and other indicators in the leachate meet the requirements of Class I standard of GB8978-1996 "Integrated Wastewater Discharge Standard". Gao et al.(2019) used cement, fly ash, and ODCPRs as raw materials to prepare subgrade materials, and the recommended dosage of cement and fly ash was 4% and 20% respectively. Cao et al.(2023) found that under the conditions of a composite solidifying agent ratio (cement to fly ash mass ratio) of 3:2, a composite solidifying agent to pyrolysis residue mass ratio of 3:2, a liquid binder to solid phase mass ratio of 0.15 to 0.20, and a curing age of 7 days, the subgrade material produced had the best compressive strength, reaching 2.77 MPa. Tan et al.(2020) used ODCPRs instead of fine aggregate in cement concrete pavement, considering strength, stiffness, and crack resistance, the optimal mix ratio of cement: ODCPRs: crushed stone: sand was 7:20:52:21. Hu (2020) and Li (2018) used ODCPRs as cement stabilized soil subgrade fill material and cement stabilized crushed stone base fill material, and the fill material performance can meet the relevant national standards. Li et al.(2023) used ODCPRs to prepare subgrade materials and simulated the leaching characteristics of pollutants under extremely acidic conditions through semi-dynamic leaching tests, and discussed the release laws and characteristics of pollutants such as naphthalene, anthracene, Cd, Zn, etc., providing important basis for the resource utilization of ODCPRs. Ran et al.(2023) found that using ODCPRs to partially replace cement in stabilized crushed stone base can enhance the mechanical properties and toughness of the material, and the reasonable replacement ratio is 10%~30%.

The advantages of using ODCPRs residues as road fill are simple process, and the key technology is to meet the requirements of bearing ratio, unconfined compressive strength, plasticity index, liquid limit, and other parameters, while avoiding heavy metal ions, benzo(a)pyrene, petroleum products, and other pollutants causing environmental pollution.

3. Used for cement production

The mineral composition of oil-based drilling cuttings ODCPRs is mainly silicate, such as quartz, limestone, and dolomite, which are also the main components of cement production raw materials. Bernardo et al.(2008) conducted industrial-scale experiments and proved that drilling waste and electric arc furnace slag can replace part of limestone and clay in cement clinker production. Zou et al.(2022) found that when the amount of ODCPRs added was less than 3.0%, the chemical composition of cement was basically stable, and the fineness, specific surface area, strength, initial setting time, final setting time, and other indicators of cement met the relevant standards. Hu et al.(2021) found that when the amount of ODCPRs added was less than 3%, the radioactivity index of cement met the requirements of "Limits for Radioactive Nuclides in Building Materials" (GB6566-2010). Xiao et al.(2023) used ODCPRs as auxiliary cementitious materials to prepare cement. Under the optimal mix ratio (clinker:gypsum: ODCPRs:slag = 71:4:7.5:17.5), the performance of the composite cementitious materials met the requirements of GB 175-2007 "General Portland Cement" P·C 32.5R grade. Liu et al. (2023) found that the addition of ODCPRs can reduce the calcination temperature of cement raw materials, and the physical and chemical indicators of cement products meet Chinese standards.

In summary, the use of ODCPRs to replace some raw materials in cement production is an important way to utilize ODCPRs. This technology has low cost, can solidify heavy metal elements, and does not affect the quality of cement products. It should be noted that the treatment of exhaust gas should be emphasized during the production process.

4. Used for ceramsite production

Sintered ceramsite are artificial lightweight aggregates made from natural resources or solid waste as the main raw materials, combined with binders such as clay, mixed with water, and granulated into spheres, and then sintered at high temperature. They have a relatively compact structure, low porosity, and good strength. Xu et al.(2023) found that with the increase of sintering temperature, the density of the ceramsite first increases and then decreases slightly. The optimal process parameters for sintering concrete ceramsite using ODCPRs are a sintering temperature of 1025°C and a sintering time of 15 minutes. Wang et al.(2023) found that when the content of ODCPRs is 50%, the compressive strength of the ceramsite is 6.31 MPa, the bulk density is 575.11kg · m^3 , the apparent density is 1097.24 kg · m^3, and the water absorption rate is 1.89%.

Using ODCPRs to prepare both non-sintered ceramsite and sintered ceramsite can meet the requirements of building ceramsite. The preparation of non-sintered ceramsite has low energy consumption and mainly relies on solidification to form spheres. It has not undergone high-temperature treatment, and there is still a potential risk of leaching of organic compounds and other pollutants in the ODCPRs. The strength of sintered ceramsite prepared from ODCPRs is relatively high, but the energy consumption is also relatively high.

5. Used for wall materials production

The chemical composition of ODCPRs is mainly composed of calcium, silicon, and aluminum, suitable for the production of masonry materials such as bricks and concrete blocks. Wang et al.(2017) demonstrated the feasibility of using ODCPRs to prepare sintered bricks and non-sintered bricks. Research by Wang et al.(2022) indicated that under the conditions of ODCPRs, cement, fine aggregate, coarse aggregate, gypsum, and polyvinyl alcohol mass fractions of 20.0%, 20.0%, 47.5%, 8.0%, 4.0%, 0.5%, and a water-solid (mass) ratio of 14%, the produced non-fired bricks can meet the MU15 strength grade specified in the "Non-fired Waste Residue Bricks" (JC/T 422-2007). By using ODCPRs as an additive material to prepare non-sintered bricks, it can replace some fine aggregates, addressing the end disposal issue of ODCPRs. Liu et al.(2021) explored the optimal process for preparing non-sintered bricks using ODCPRs, with ODCPRs mass fractions reaching up to 40%. It was confirmed that the silicon, calcium, and aluminum components in ODCPRs participate in the sintering reaction in the system, promoting the formation of glass phase in the system, filling internal pores, and aiding in the formation of dense sintered brick bodies. Wang et al.(2017) found that ODCPRs and fly ash as substitutes for fine aggregate and cementitious materials in concrete are feasible. When the amount of ODCPRs is 35%, the best workability and compressive strength can be obtained. Mohammed et al.(2011) discovered that ODCPRs can be used to replace sand and stone for preparing concrete blocks, with compressive strength comparable to ordinary concrete blocks, higher density, lower water absorption rate, and thermal conductivity.

In conclusion, using ODCPRs as a raw material to produce non-sintered bricks, sintered bricks, concrete blocks, and other materials is technically feasible. A high addition of ODCPRs is beneficial for the large-scale utilization of ODCPRs.

6. Conclusion

(1) ODCPRs can be used as road fill, meeting the relevant requirements of Chinese standards for bearing ratio, unconfined compressive strength, plasticity index, liquid limit, etc. During usage, attention should be paid to avoiding heavy metal ions, benzo(a)pyrene, petroleum, and other pollutants that may cause environmental contamination.

(2) ODCPRs can be used as a raw material to produce non-sintered ceramsite, non-sintered bricks, concrete blocks, and other building materials. The production process is simple and does not undergo high-temperature treatment. However, there still exists a potential risk of leaching from organic pollutants and other contaminant components in ODCPRs, highlighting the importance of the stability of pollutant solidification effects.

(3) Sintered ceramsite and sintered bricks are formed through high-temperature calcination, resulting in low environmental pollution risks in the later stage. Nevertheless, efforts should be made to strengthen the control and management of production waste gas during the production process.

(4) Using ODCPRs to prepare cement and wall materials does not yield high product added value. Therefore, continuous efforts should be made to develop high-value-added products and broaden the pathways for the high-value utilization of ODCPRs.

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