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Review

Analysis and recommendations on the use of polymer and phenol-based materials for coal mines

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A B S T R A C T

In this study, besides summarizing the properties of polymer materials used in underground coal mines, the problems that may arise in the use of polymer materials used in coal mines and the deficiencies of the relevant existing standards are discussed. Most of the safety accidents of polymer materials are encountered with results such as spontaneous combustion of materials, combustion of coal seams due to these combustions, and the release of toxic and harmful gases. The main causes of combustion accidents can be summarized as high reaction temperature, insufficient flame retardant, uncontrollable reaction process, defective standards and awareness of insufficient safety materials. Within the scope of this study, health problems that may be encountered due to the carcinogenic formaldehyde content of phenol-based filling materials will also be mentioned, and reasonable suggestions and measures will be discussed to prevent these and similar situations. The aim of this study is to establish a guide on the safe use of polymer materials in underground coal mines and correct understanding of the possible risk associated.

Keywords: *Polymer materials, Underground coal mines, Safety precautions, Spontaneous combustion, Reaction temperature, Flame tightness.*

Introduction

Polymer materials are widely used in coal mines and there are many types including polyurethane, epoxy resins, urea-formaldehyde resins for the main problems related to ventilation/leakage or cracks of coals (Liu 2021). In addition to that, some sodium silicate/polymer composite gels are recently being used for the prevention of coal spontaneous combustion (Ren et al. 2019). The polymers are mostly needed in terms of the isolation purposes and they are desired to show high strength, low reaction temperatures and no harm to human health. The scope of the application of phenol based filling materials in underground mining is summarized as in the following: consolidation of the structure of the weak coal strata to be produced or excavated, filling the cavity between steel support and ground, goaf stabilization, sealing of all entries connected to the fire area and isolating coal surface in the fire zone or area of spontaneous combustion. Accordingly, purposes of the usage of these abovementioned phenol based filling materials are provided as in the following:

i. Cavity filling

ii. Air-tight ventilation seals which will aid underground ventilation requirements

iii. Isolate the coal from air flow

iv. Fighting underground fires by constructing airtight barrier to cut off oxygen supply

The characteristics of the polymer materials have been summarized by Liu 2021 which are "high infiltration permeability, fast setting speed, short curing time, small shrinkage after grouting, good bonding and compressive properties, strong durability and impermeability". As well as the need for these polymers in mines increases day by day, the production statistics was reported as 40 000-50 000 tons/year by Liu 2021, for the coal mines (more than 2000 mines) in China. Keeping in mind the abovementioned statistics by Liu 2021, need for the polymers (specifically phenol based filling materials) in coal mines in Turkey is recently increased almost 30%-50% and the respective amounts for year 2019 and 2020 were reported as in Table 1 (Anon 2021).

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Table 1. *Usage of phenol based filling materials in Turkey in 2019 and in 2020 (Anon 2021).*

REGION	Total amount of usage in 2019 (tons)	Total amount of usage in 2020 (tons)
SOMA	1650	2880
ANKARA	300	400
ZONGULDAK	160	210
OTHERS	30	45
TOTAL	2140	3535

In addition, Çavuşoglu (2008) has a study which investigates the potential use of fly ash as a backfill material for the specific mine in Çayırhan-Turkey. Although the study abovementioned does not refer to the polymers or phenol based filling materials, the need for these materials in underground mine environments can be very well understood with this study. Zhang and Sun (2019) have carried out experiments and investigated mechanism of a polymer based material for reinforcing purposes of broken coal mass. In their study of Zhang and Sun (2019), authors have widely described the fractures, as primary and ongoing. They (Zhang and Sun 2019) have stated the fact that primary fractures result in expansion and transfix which initiates large number of initiated secondarily fractures, even as worse as coal rupture and many problems related to mechanical properties of coal mass (Shi et al. 2017, Wang and Yang 2017, Zhang and Sun 2019). Referring back to Zhang and Sun 2019 study, they have stated the fact that stress state changes for newly exposed coal seams and authors have expressed this as a reason of the damages of coal mass and rapid reduction of its bearing capacity. The importance of initial fractures and crack initiation is mainly because of the results abovementioned and the risks of inducing coal gas outburst disasters due to the result due to ineffective resist of the ground stress and gas expansibility (Zhang and Sun 2019).

Polymers, phenol based filling materials in specific has their significance based on the air leakage/ventilation problems which also has a vital role in underground mines. Phenol-based fillers are widely used in mining, and especially in underground coal mining, as an effective material for filling and blocking air leaks due to its heat resistance, flame retardancy, good sealing and ease of construction (Lei et al 2010, Wang et al. 2019). In addition to the usage in underground coal mining, Lei et al. (2010) have summarized their usage for civil construction, passenger and military aircrafts, marines and electronic applications. In Figure 1, a representative of phenol formaldehyde polymer condensation reaction is provided. The SEM microphotographs of the phenolic foam (Lei et al. 2010) is provided in Figure 2.

In addition, Ren et al. (2019) have investigated the novel sodium silicate/polymer composite gels and authors have suggested the usage of this polymer based material in order to prevent spontaneous combustion of coal. In their study of Ren et al. (2019), the authors have given information about the attempts of the researchers to prevent spontaneous combustion of coal by applying different technical measures and materials, up to year 2019. Ren et al. (2019) have cited the studies (Qin et al. 2015, Lu and Qin 2015) which includes pumping the top coal caving regions of coal drifts with foam cement, and some other papers (Hu and Wang 2013, Hu et al. 2014A, Hu et al. 2014B) which includes filling them with a polymeric material to seal air leakage. In the same context as in the Ren et al. (2019) study.

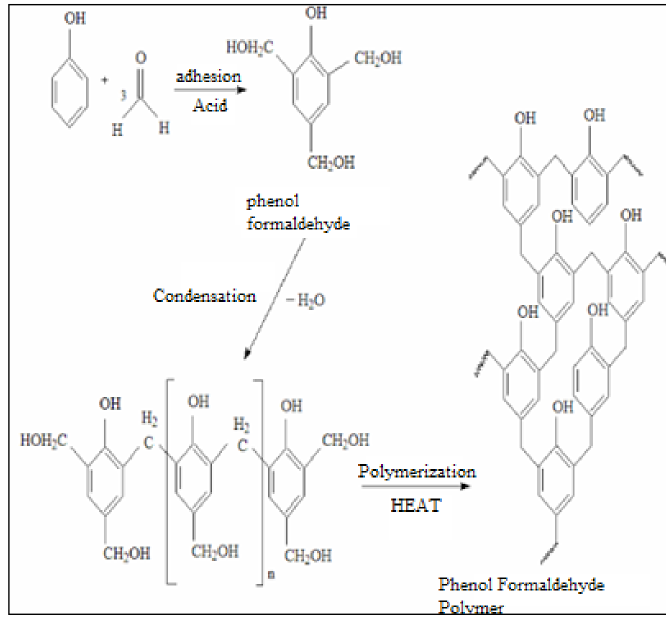

Figure 1. *Phenol formaldehyde condensation reaction (Frihart, 2005, Özlüsoylu 2016).*

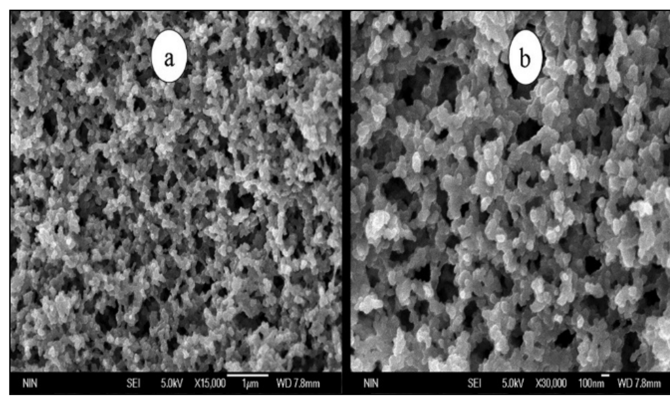

Figure 2. *The SEM microphotographs of the phenolic foam (adapted from the study of Lei et al. 2010)*

Fan et al. (2020) have proposed a novel plastogel which was claimed to have the ability to prevent and control fires in coal mines. The plastogel proposed by Fan et al. (2020) was described with its preparation by adding coagulant, polymer plasticizer and bentonite into the water glass solution. With the understanding of the abovementioned papers, it can be clearly emphasized the fact that polymer materials have been widely used in underground coal mining applications mainly for the purposes of sealing the air leakage and prevention of spontaneous combustion. In addition, these abovementioned polymer materials (with one, two or even three components (like plastogel by Fan et al. 2020)), should be tested in terms of their possible hazardous composition.

Güner and Öztürk (2019) have investigated deformability behavior of thin spray-on liners both experimentally and numerically. Although TSLs (thin spray-on liners) will not be detailed in this study, still they are polymeric or cement-based products that have been used for underground mine operations extensively for the purposes of rock support. Being produced based on the polymeric materials, TSLs should also be reconsidered and examined in terms of their hazardous content as regards to occupational health and safety.

Formaldehyde is one of these hazardous components of some of these polymer materials which are widely utilized in underground coal mines. In order to understand possible formaldehyde exposure, circumstances of exposure and results accordingly, report by AWES (2014) can be suggested to be a guide in the first place. As regards to USA-OSHA (Occupation Safety and Health Administration) standards formaldehyde concentration of gas produced by the reaction between resin and catalyst should be limited to 2 ppm.

In this study, polymers and phenol based filling materials were investigated in terms of the accidents caused by their utilization in underground coal mines. In addition, some examples of the accidents were provided in this context with their analysis. Not only the accidents based on the usage of these polymer materials and phenol based filling materials were examined but also some suggestions were provided by means of occupational health and safety.

1. Analysis and reasons behind the accidents resulting with polymer materials utilization in underground coal mines

Since chemical reaction is undergoing between the composites of the polymer & phenol based filling materials, each composite, final product and the chemical reaction should be taken into consideration all together in order to correctly address the hazardous structure of the specific material. To be clearer, -in the case the composites A and B forming polymer C, not only polymer formed (C) should be tested but also A and B along with the chemical reaction should be investigated in terms of their hazardous nature in specific atmosphere such as underground coal mining. Since underground coal mining is not directly in contact with the atmosphere, polymer materials (C), composites (A, B) and chemical reaction (between A and B) might be dangerous and hazardous in specific atmosphere of underground coal mining. Flash point temperature of each composite (A and B) should not be low, and it should be the first issue raised in underground coal mining application. The reason is rule of thumb, if the flash point temperature of the composites are lower than reaction temperature, that would result in fires that can trigger spontaneous combustion of coal. If the flash point is too high, then the polymer product is not classified as flame resistant. According to Liu (2021), polymeric materials and their composites along with the chemical reaction in between may cause carbonization failure, spontaneous combustion, explosion, fire, combustion, and toxic and harmful gases lead to poisoning and corrosion accidents. According to Küçük and Ilgaz (2015), accident causes by technical reasons (Güyagüler and Bozkurt 1993, Akkaya 2001, URL-1 2014, Durşen and Yasun 2012) were summarized as in the following:

1. Accidents caused by electricity and mechanization systems failures.

2. Explosions caused by dust, gas and radiation.

3. Explosives usages in mining operations.

4. Collapses and strata failures.

5. Haulage and water drainage system malfunctioning reasoned accidents and flooding.

6. Open pit fires, self-heating of coal related problems.

7. Sudden inrush, spontaneous combustion, pressurized gas discharges.

8. Accidents during the operations of coal preparation & mineral processing.

9. Low quality of mechanical maintenance, laboring and operational misguidances.

10. Accidents observed during the transportation, preparation, and usage of materials and instrumental tools.

11. Accidents due to mine environment constraints (heat, moisture, pressure, steam, noise, lighting, sliding surface, etc.).

12. Personnel and occupational health & safety related problems (ability, education, motivation, physical and mental state, personal attention, personal protection, etc.).

13. No proper working conditions and working environment (Küçük and Ilgaz 2015).

2. Examples of accidents and their causes

According to Liu 2021, a fire accident occurred in four mining areas (4238 fully mechanized working face in + 1030 m level) of a coal mine in Sichuan Province. This fire accident abovementioned occurred during the process of using organic polymer filling reinforcement material to deal with the top high caving area of the specified support, and it resulted in the closure of the whole mining area (Liu 2021). According to Liu 2021, direct causes of this abovementioned fire accident are: "rise in the internal temperature of the material due to heat release caused by the fire, ignition of wood stack, ignition of coal wall and return air side combustible and coal seam, fire". The indirect causes of the accident are stated as: "(i) Security technology management confusion. (ii) Mixing different polymer materials. (iii) Poor air circulation environment. Since the front is a closed falling space, which is almost not circulated, and the volume of the falling space is limited, a large amount of heat cannot be, forming a heat accumulation area. (iv) Spontaneous combustion of materials. Water injection cooling is not enough, the head of the water and water pressure is not large, can not inject a large number of cooling water at high temperature point, resulting in the combustion of polymer material itself, and then cause coal combustion" (Liu 2021). Not only accidents in terms of polymer based materials for the purposes of support and strata in mines but also gas inhaling and smoke based accident was observed in China, in specific mines (Liu 2021) observed. According to Liu (2021), toxic and harmful gases resulted in the death of a worker who was objected to the "yellow smoke" at the bottom of the drilling site door. Harmful gases were released by the chemical reaction of polymer materials filled in drilling site in a coal mine in Huaibei (5 # drilling site of 7118 working face). According to Liu (2021), direct causes of the accident is summarized as: "the filling material is not qualified, after filling the high temperature and toxic and harmful gas caused by poisoning death". Liu (2021) has also mentioned about the indirect causes of the abovementioned toxic and harmful gases' accident as: "(i) The procurement of filling materials is random. Not in accordance with the provisions of the company, from the normal channels of regular manufacturers import, acceptance. (ii) The material acceptance personnel in the ventilation area failed to work, only received quantity and did not check the quality. (iii) Poor hazard identification ability. Ventilation area gas inspectors in the third inspection smell, see yellow smoke but not informed. (iv) Unclear job responsibilities. Institution adjustment, the outburst prevention area has just been listed and has not been properly operated; The construction personnel directly used the filling material without identification, resulting in the chemical reaction of the filling material and producing high-level and toxic and harmful gases" (Liu 2021).

3. Problem in current standards and technical uncertainties

Usage of polymer materials in coal mines were issued by four standards which are being implemented since 2011 in China (Liu 2021). Existing situation about the application of these abovementioned standards is regarded as "not ideal" by Liu (2021) and yet it is also mentioned the fact that there are great objections to them in China (Liu 2021). By referring back and forth to Liu (2021), current standards in China were stated to only regulate the flame retardancy, antistatic ability and mechanical properties of polymer materials under normal temperature conditions. This abovementioned criticisms about the standards and their corresponding regulations are also valid for the ones which are being implemented in Turkey (See the example of a technical requirements of phenol based filling materials, Table 2)

Table 2. *Technical requirements of phenol based filling materials in Turkey.*

Technical Requirement	**Explanation**
Flame Retardancy	The foam will be flame resistant (Flame Retardant).
Reaction Temperature	The reaction temperature will be < 90°C.
Antistatic Property	The foam will be antistatic (≤109Ω).
Strength	Compressive strength shall not be less than min 0.02 MPa at 10% stacking.
Free Formaldehyde Content	The amount of free formaldehyde of the resin component used for foam formation will be less than 0.1% by weight.
Swelling Rate and Reaction Rate	High expansion rate and instant (immediate foaming) reaction.
Availability for Fire Fighting	It will be suitable for fire fighting activities.
Resistancy for Degradation in underground atmosphere	The foam formed as a result of the reaction is resistant to underground weather conditions, water, solution and biological disturbances.
Flash and Ignition Point	Low reaction temperature that will not self-ignite. The flammability of the reaction heat should be lower than the flash point temperature of the resin and catalyst used in the formation of the foam.
Blocker Property for Coal Spontaneous Combustion	It will stop the spontanous combustion of coal. It will stop the heating process of coal by isolating coal from air flow.
Long-term Availability of the Foam Structure	The stability of the foam structure will not change in the long term under underground conditions.
No Possible Effect on the Surroundings (sensors, etc) in Underground Atmosphere	No effect of foam application on the sensors in the underground atmosphere.
Property for Occupational Health and Safety Regulations	It should be proper for occupational health and safety regulation.

Liu (2021) have widely described the main problems in the standards (standards in China) of either for reinforcement and water plugging polymer materials (Table 3) and filling and sealing polymer materials (Table 4).

Table 3. *Main problems in the standards (standards in China) for reinforcement and water plugging polymer materials (Liu 2021).*

Problem in Standard	Problem Description
(1)	In AQ1089 - 2011, according to the different parts of reinforcement materials in coal and rock mass, it can be divided into C (coal reinforcement) and R (rock reinforcement). In the practical application process, it is difficult to distinguish in most cases.
(2)	The anti-aging performance index is expressed as 'no change in surface and no loss in mass', which is problematic. In the aging process, the volatilization of some solvent molecules will reduce their mass, but generally will not affect their mechanical properties.
(3)	The expression of " hazardous substance limit " in the standard is relatively vague. It should be concretized according to the material type, increase the flue gas toxicity test of the material, and quantitatively detect the content of toxic and harmful gases (HCN, NO, CO, halogen acid gas, etc.)

Table 4. *Main problems in the standards (standards in China) for filling and sealing polymer materials (Liu 2021).*

Problem in Standard	Problem Description
(1)	The materials in AQ1090 - 2011 were divided into N and P categories. In the actual grouting process, the materials used for hole sealing also need to bear pressure, so there is no need to classify them.
(2)	The standard flash point determination problem, generally A material can not measure the flash point, 50 ~ 60°C began to bubble, 80 ~ 90°C overflow pot, 100 ~ 110°C condensation; Material B is an acid without flash point. Substances with low boiling points (foaming agent) may be added in component A. After heating, the gas overflows and expands, and material A will bubble, overflow pot or even condense, which is difficult to measure.
(3)	The expansion ratio specified in the standard is not less than 25 times. Due to the different underground environments of coal mines, some need large expansion ratios, and some do not. In general, the higher the expansion ratio is, the lower the mechanical strength of the material is, and the expansion ratio of the material is not less than 10 times.
(4)	The expression of 'hazardous material limit' in the standard is vague and should be specified according to the type of material; The flue gas toxicity test of materials should be added to quantitatively detect the content of toxic and harmful gases (HCN, NO, CO, halogen acid gas, etc.).

The sector of polymer materials used in mining and underground coal mining is a rapidly developing sector and there are two problems that can be associated with the rapid development of this sector. First, the research and development potential of polymer reinforcement materials is insufficient. The second is that the users of polymer materials do not have enough information about the properties of the materials (Liu 2021). When the polymer materials usage standards in mining applications in Turkey (Table 2) and in China (Table 3, Table 4) are evaluated, it is understood that the users of polymer materials are not well-informed and research and development activities are not carried out at a sufficient level. For example, when the standards in Turkey (Table 2) are evaluated, it has not been determined in which standards these polymer materials should be tested for most of these technical requirements. Similarly, in the study of Liu (2021), author described the expressions in the standards for China (Table 3 and Table 4) as "problematic", "not clear enough", "indistinguishable", "cannot be generalized and should be specified specifically for the mine", "difficult to measure" and "do not need to be classified".

Conclusions

In addition to its high risk intense structure of underground coal mining, usage of polymer materials might have potential risks in terms of their hazardous and toxic content. Due to the high demand for the polymers with their available solutions to the problems in underground coal mining, corresponding industry is developing rapidly. This rapid development of polymer industry for mining applications results in two main problems: (i) lack of knowledge about the properties of polymers, (ii) lack of research and development about the applicability of these polymers. Based on the abovementioned problems for polymer materials, accidents or health disorders resulted by short-middle-long term objection might be unfortunately observed due to their hazardous and toxic structures. Considering the availability of polymer materials for the corresponding problems in underground coal mines, usage of polymer materials is inevitable. However, safe production of coal mines should be the top priority with the proper selection of polymer materials (least harmful, best convenient). Proper selection of polymer materials should include flue gas toxicity test and the content of toxic and harmful gases (HCN, NO, CO, halogen acid gas, etc.) should be quantitatively detected. In order to evaluate the abovementioned polymer material specifications (least harmful, best convenient), more research should be conducted in this field. With the help of the current developments and research carried out, technical requirements should be revisited and reworded. In this case, standards for the evaluation of polymer materials, the preference should be made according to test results obtained previously well-defined and described experimental conditions & standards. Technical uncertainties can only be overcome with the standard testing methods and technical specifications of the polymer materials should include the description of this standard testing methodology. Since, objection of the gases from these polymer materials (either during the chemical reaction or during the application) and interrelationship between this objection (short-middle-long term) and health issues is not yet well established, occupational health and safety precautions should be taken into consideration. Reviewing the occupational health and safety precautions, least harmful alternatives should be recommended by independent authorities. Cheaper alternative of polymers will always be available while their corresponding level of toxicity is questionable. Independent authorities should evaluate the polymer materials in terms of their potential risks (hazardous, toxic content) and price of each alternative should be out of their concern.

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