Petroleum and Coal

Article **Open Access**

Research and Application of the Rehbinder Effect in Coal Technologies

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Received December 15, 2025; Accepted April 27, 2026

Abstract

This article explores how the Rehbinder effect manifests during the processing of low-metamorphosed coal from the Donetsk Basin. The authors distinguish between the classical primary Rehbinder effect (material weakening) and the secondary Rehbinder effect (a change in the zeta-potential of crushed coal) also known as the "adsorptive exposure of the pore space of the solid phase"), which accompanies the primary Rehbinder effect. The goal of the study is to examine how both primary and secondary Rehbinder effects manifest in Donetsk coal. Several methods were applied, including coal-sizing analysis, electroosmotic measurement of coal surface ζ-potential, the Tovarov method for determining the external specific surface area of dispersed material (coal), the authors' own method for preparing polished sections of coal grains and analysing oxide films within pore walls and fractures, as well as the analysis of the results using Microsoft Excel. The experiments confirmed that the primary Rehbinder effect productively weakens the organic matter of low-metamorphosed Donetsk basin coal. The paper outlines several objectives for expanding future experimental work considering the following factors: pore parameters – their size and surface characteristics, the composition of the weakening liquid infiltrating into the pores, as well as infiltration kinetics. In the coal studied, the secondary Rehbinder effect appears as a shift in the hydrophilic–hydrophobic balance of the overall external surface of the crushed coal towards increased hydrophilicity. This shift results from the exposure of oxidised pore surfaces during the primary Rehbinder effect, as these surfaces contribute their oxidised fraction to the external surface when the pre-soaked coal is crushed. The experimental data show that it is possible to produce a stable, highly concentrated water-coal slurry (water-coal fuel) from the low-metamorphosed Donetsk coal at pH > 4 (ζ > 30 mV) when the coal is pre-soaked and at pH > 6.7 when dry-ground.

Keywords: *Rehbinder effect; Crushing, sizing analysis; Highly concentrated water-coal slurry; Zeta-potential; Tovarov method; Oxidized film; Osmotic method.*

1. Introduction

The classical interpretation of the Rehbinder effect defines it as adsorptive strength reduction - a modification of the mechanical properties of solids driven by physical and chemical processes that lower the body's surface (interfacial) energy, which can ultimately facilitate deformation and fracture [1-3].

Contemporary physical and chemical interpretation views the Rehbinder effect as a stress-dependent adsorption phenomenon in which surface-active agents weaken solids by reducing surface energy, loosening atomic bonds, promoting dislocation mobility and chemically assisting fracture propagation. The key feature of this synergetic mechanochemical process is predominantly non-chemical interaction between the solid and its environment along internal fracture surfaces. However, the deformation of interatomic bonds within a solid (including

coal) when its atoms interact with adsorption-active components can substantially reduce both the work of plastic deformation and the specific fracture energy overall [4-5]. Notably, such modifications of interatomic bonding in the oil-treated coal matter have been confirmed by our X-ray structure analysis [6]. A simplified representation of solid fracture via the Rehbinder effect is shown in Fig. 1.

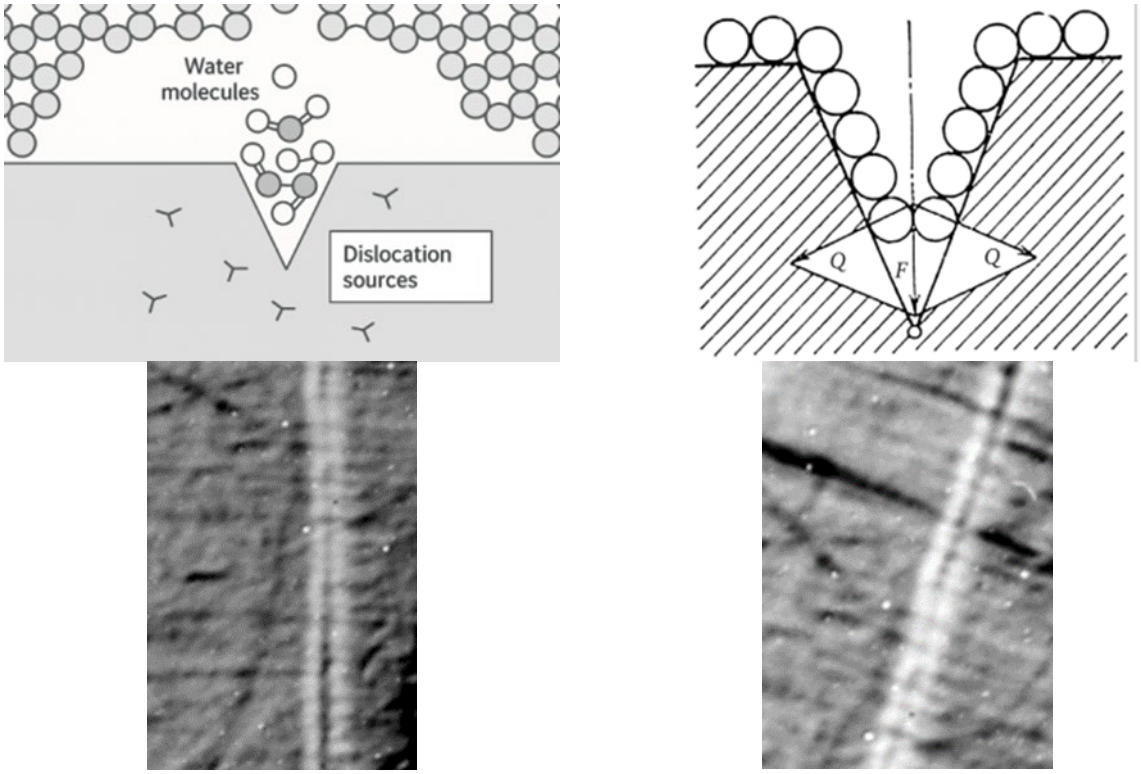


Figure 1. Diagram of the adsorption-wedging effect of polar water molecules (a, b) on hydrophilic oxidised pore and fracture surfaces (c, d – white oxide films on pore walls in coal grains, authors' own photographs of polished sections, x650).

Ongoing research into the Rehbinder effect has revealed additional consequences of its manifestation. While developing the theoretical foundations of highly concentrated water-coal slurries (water-coal fuel, or WCF), a team of Ukrainian researchers proposed a hypothesis that the behaviour of WCF could be interpreted within the framework of lyophobic colloids. To assess the conditions for WCF stability, they applied the Derjaguin–Landau–Verwey–Overbeek theory of lyophobic colloid stability (DLVO theory) [7]. These studies identified a shift in the hydrophilic-hydrophobic balance of the overall external surface of crushed coal, occurring with the primary Rehbinder effect (material weakening). The secondary Rehbinder effect (the change in zeta-potential of the crushed coal) was termed the "adsorptive exposure of the pore space of the solid phase" (documented by VS. Biletskyi *et al.*, 2012-2014) [8-10].

The need for fine and ultrafine coal has several technological processes, including mineral processing, to expose mineral inclusions in the raw coal (flotation, flocculation, oil agglomeration [11-12]), coal briquetting [13-14], pyrolysis, gasification and chemical processing — ultrafine coal is a key component in processes such as hydrogenation (H-coal, synthetic oil), low-temperature carbonisation, catalytic synthesis of carbon materials, and the production of sorbents and activated coal [15-17].

Both the primary and secondary Rehbinder effects play an important role in the formulation of water-coal fuel [7,18]. The Rehbinder effect is also likely to operate during hydraulic fracturing (fracing) of coal seams and the adjoining rock in extraction and degasification [19-20].

The goal of the study is to investigate how the primary and secondary Rehbinder effects manifest in low-metamorphosed coal from Donetsk basin. To achieve this goal, the study addresses the following objectives:

- to determine the parameters governing the primary Rehbinder effect during the crushing of low-metamorphosed Donetsk coal;
- to refine the conditions under which the secondary Rehbinder effect can be utilised in producing a highly concentrated water-coal fuel slurry based on low-metamorphosed Donetsk coal.

2. Experimental (Methods and materials)

The research was conducted using low-metamorphosed coal from Donetsk basin with an ash content of A^d = 16.5%. The sizing analysis of the raw coal is shown in Table 1.

Table 1. Particle size distribution of the raw coal.

Size fractions, mm	Fraction yield, γ, %	Total fraction yield, Σγ, %
>3.0	8.9	8.9
1.0-3.0	19.65	28.55
0.5-1.0	18.1	46.65
0.2-0.5	15.5	62.15
0.1-0.2	5.0	67.15
0.08-0.1	4.95	72.109
0-0.08	27.9	100

Two crushing approaches were assessed: crushing in an industrial ball mill after pre-soaking the coal in an ad-hoc screw mixer-feeder to a solids content of 62%, and crushing without pre-soaking. Both crushing processes were performed at the same mill throughput. During pre-soaking, the plasticiser "Dofen" was added into the water at 1% of the dry coal mass. Coal-sizing analysis was conducted using the vibrating sieve analyser shown in Fig. 2.

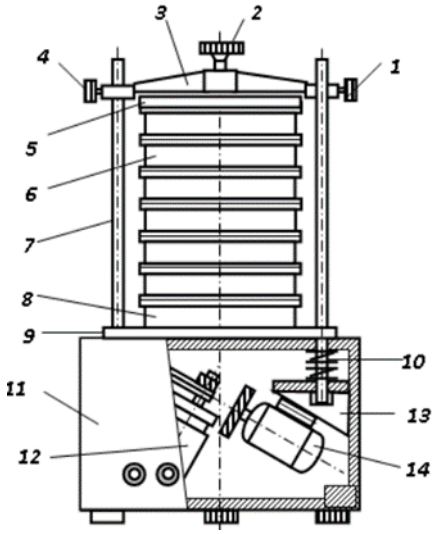


Figure 2. Vibrating Sieve Analyser: 1, 2, 4 – fastening screws; 3 – crossbeam; 5 – lid; 6 – set of sieves; 7 – pylons; 8 – tray; 9 – support; 10 – springs; 11 – housing; 12, 14 – electric motors; 13 – brackets.

A new technique developed by the authors was employed to prepare polished sections of coal grains and to analyse the presence of oxide films on pore walls [21-22].

The external specific surface area of both the raw and crushed coal was measured using the Tovarov method [23-24]. The version of the Tovarov apparatus used, designed by the National Technical University "Kharkiv Polytechnic Institute", is shown in Fig. 3. The zeta-potential of the coal was determined using a device developed at Donetsk National University (Yu.L. Papushyna) [25], as shown in Fig. 4.



Figure 3. The Tovarov apparatus was employed in the experiments. design variant of the Department of Oil, Gas and Condensate Production, NTU "Kharkiv Polytechnic Institute".

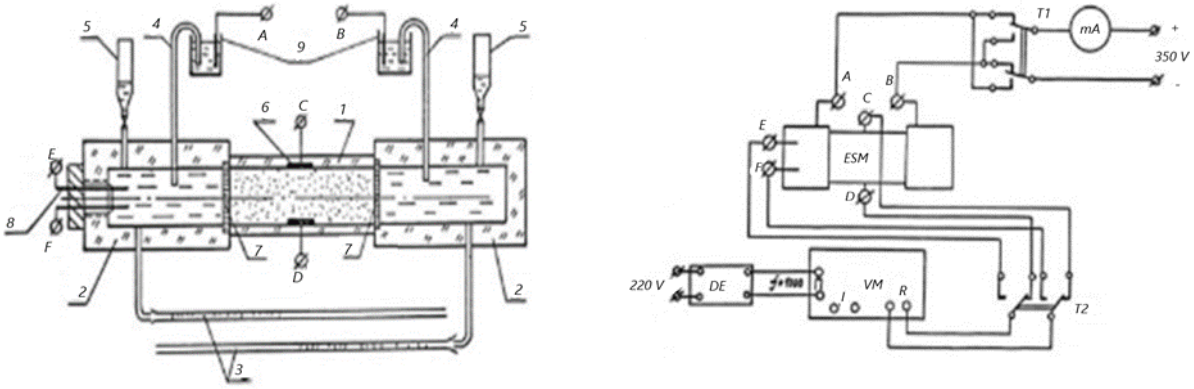


Figure 4. Electroosmotic setup for measuring zeta-potential. A – diagram of the device and B – diagram of its external electrical connections. 1 — removable vessel for the test material; 2 — side chambers; 3 — graduated capillaries; 4 — electrolytic agar bridges; 5 — auxiliary burettes; 6 — flat platinum electrodes; 7 — removable filters; 8 — platinum rod electrodes; 9 — electrolyte vessels with copper electrodes.

3. Results and discussion

3.1. Investigation of the primary Rehbinder effect (coal weakening prior to crushing)

Particle size distribution of coal ground in dry and pre-soaked conditions is summarised in Table 2. To facilitate analysis, the results were visualised using Microsoft Excel. Figure 5 shows the total yields Σγ, % for each size fraction of the raw coal and of coal after ball-mill crushing under both dry and pre-soaked conditions. Figure 6 presents yields γ, % for corresponding

size fractions of the raw coal and of coal after ball-mill crushing under both dry and pre-soaked conditions.

Table 2. Particle size distribution of crushed coal.

Size Fractions, mm	Without pre-soaking		With pre-soaking	
	Fraction yield, γ, %	Total fraction yield, Σγ, %	Fraction yield, γ, %	Total fraction yield, Σγ, %
0.30-0.50	1.86	1.86	0.5	0.5
0.25-0.30	4.37	6.23	2.5	36.9
0.10-0.25	18.12	24.35	8.8	11.8
0.08-0.10	15.29	39.64	19.2	31.0
0.04-0.08	7.44	47.08	5.9	36.9
0-0.04	52.92	100	63.1	100

Analysis of the tabulated data and curves of total yields by size fraction Σγ, % for raw coal and coal after ball-mill crushing under both dry and pre-soaking conditions reveals a significant increase in the yield of the 0.08–0.10 mm and 0–0.04 mm fractions when the coal was pre-soaked. Notably, the curves plotted on a semi-logarithmic scale proved more indicative than those on a linear scale (see Fig. 5).

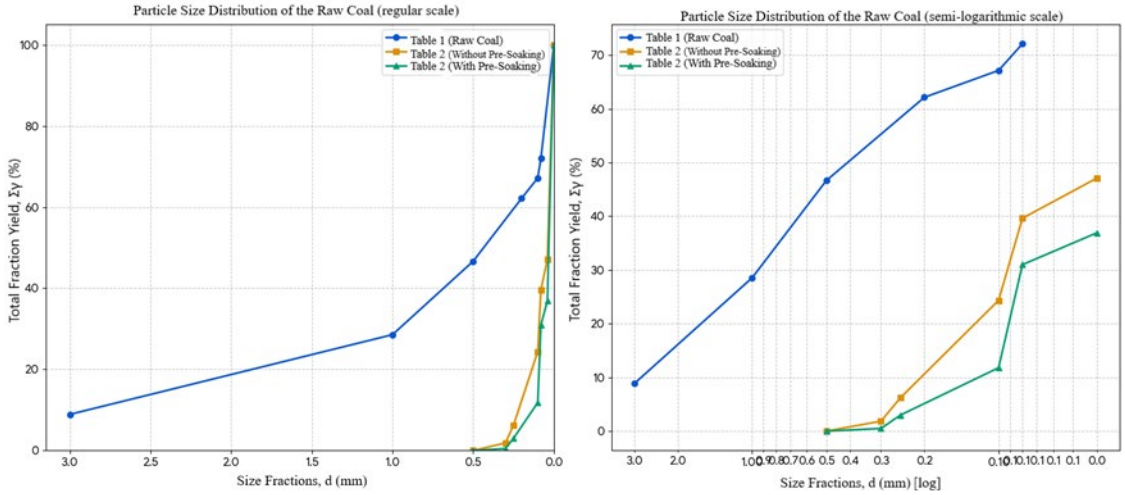


Figure 5. Total yields by size fraction σγ, % for raw coal and coal after ball-mill crushing under dry and pre-soaked conditions. Left – linear scale, right – semi-logarithmic scale.

This same trend of improved ball-mill crushing under pre-soaking conditions is also evident in the yields γ, % by size fractions for raw coal and ball-milled coal with and without pre-soaking. Processing the results in Microsoft Excel using different display modes, such as 4a – standard; 4b – smoothed curves; 4c – with filled areas, provides a more detailed view of how individual size fractions change during crushing (see Fig. 6).

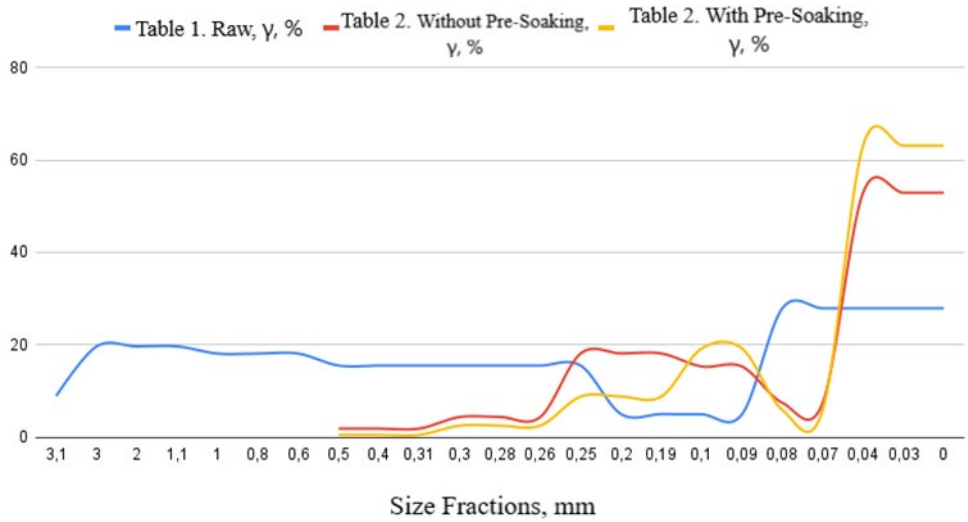
Additionally, using the Tovarov method to assess the grain material's size allowed an integrated evaluation of the Rehbinder effect when soaking crushed coal. The results, summarised in Table 3, confirm that pre-soaking has a positive impact on coal comminution.

Table 3. External specific surface area of coal determined using the Tovarov method.

Parameter	Raw coal	Crushed coal without pre-soaking	Crushed coal with pre-soaking
External specific surface area of coal, $S_{z.p.}$, cm^2/g	1723	2952	3570



a



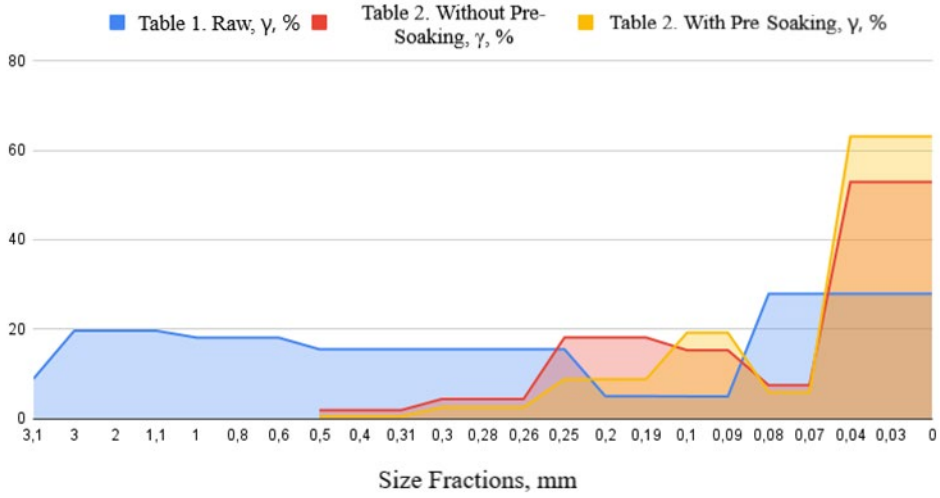


Figure 6. Yields γ, % by size fractions for raw coal and ball-milled coal with and without pre-soaking: 6a – standard; 6b – smoothed curves; 6c – with filled areas.

The external specific surface area $S_{z.p.}$ of coal crushed after pre-soaking was 1.21 times greater than that of coal crushed under the same conditions without pre-soaking. These results experimentally confirm that the Rehbinder effect productively weakens the organic matter of low-metamorphosed coal from Donetsk basin under the conditions studied. However, to quantitatively evaluate and predict the comminution results for organic matter across different coal metamorphism grades, the experimental framework needs to be expanded to account for several factors.

First, the impact of the Rehbinder effect strongly depends on the characteristics of pores and fractures in the coal material exposed by liquid films. As shown in [26], pore parameters are influenced by the degree of coalification. Second, the infiltration of liquid (water+reagents) into coal pores and fractures is affected by the hydrophilic-hydrophobic properties of the pore surfaces, as well as by the composition and characteristics of the infiltrating liquid and any reagents used. Third, the study [27] has shown that complete filling of coal pores and fractures with water can take 25–30 days. The effective soaking time (during which the weakening effect of the Rehbinder mechanism is significant), seems to depend on the dynamics of liquid phase infiltration, which remains to be investigated.

3.2. Investigation of the secondary Rehbinder effect (changes in the hydrophilic-hydrophobic balance of external surface of the weakened and crushed coal)

To address the second research objective, i.e. to refine the conditions under which the secondary Rehbinder effect can be utilised in producing highly concentrated water-coal fuel slurry based on low-metamorphosed Donetsk coal, we measured the zeta-potential of the crushed coal. This is motivated by previous studies on the sedimentation stability of highly concentrated water-coal slurries, which based on the DLVO theory, demonstrated that the overall potential φ and the zeta-potential ζ of coal particles are key factors controlling the slurry's stability. Specifically, our previous work [28] showed that for $\varphi < 50$ mV, mineral slurries are aggregately unstable. Its particles irreversibly coagulate due to Van Der Waals dispersion interactions, resulting in fractioning of the mineral slurries. When $\varphi > 50$ mV, the repulsive energy barrier increases, thereby enhancing the aggregate stability of the mineral fraction in the water-coal slurry. As reported in [29], the ζ-potential has a critical role in the stability of coal-water slurries, which reflects the electrostatic repulsion between coal particles. A high absolute zeta-potential, typically above (30) mV or below (-30) mV, generates strong repulsive forces that prevent particle aggregation and promote a stable, well-dispersed slurry. Conversely, a low zeta-potential produces weak repulsion, allowing particles to adhere and settle under Van der Waals forces.

The experimental zeta-potential data for crushed coal, pH = 1–11, 5–8, are presented in Tables 4 and 5, and Figures 7 and 8.

Table 4. Zeta-potential of crushed coal, pH = 1–11

Without pre-soaking prior to crushing		With pre-soaking prior to crushing	
Zeta-potential, mV	pH	Zeta-potential, mV	pH
0	1	0	1
5	2	10	2
9	3	23	3
19	5	37	5
25	6	44	6
32	7	55	7
39	8	60	8
60	11		

Table 5. Zeta-potential of crushed coal, pH = 5-8.

Without pre-soaking prior to crushing		with pre-soaking prior to crushing	
Zeta-potential, mV	pH	Zeta-potential, mV	pH
19	5	37	5
25	6	44	6
32	7	55	7
39	8	60	8

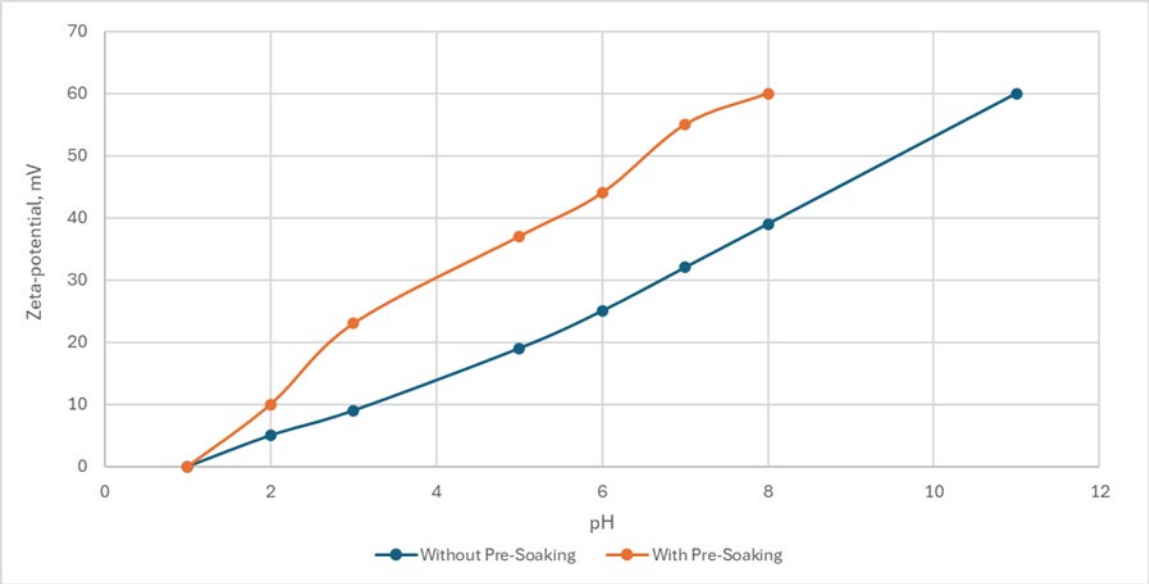


Figure 7. Experimental Zeta-potential data for crushed coal, pH = 1–11.

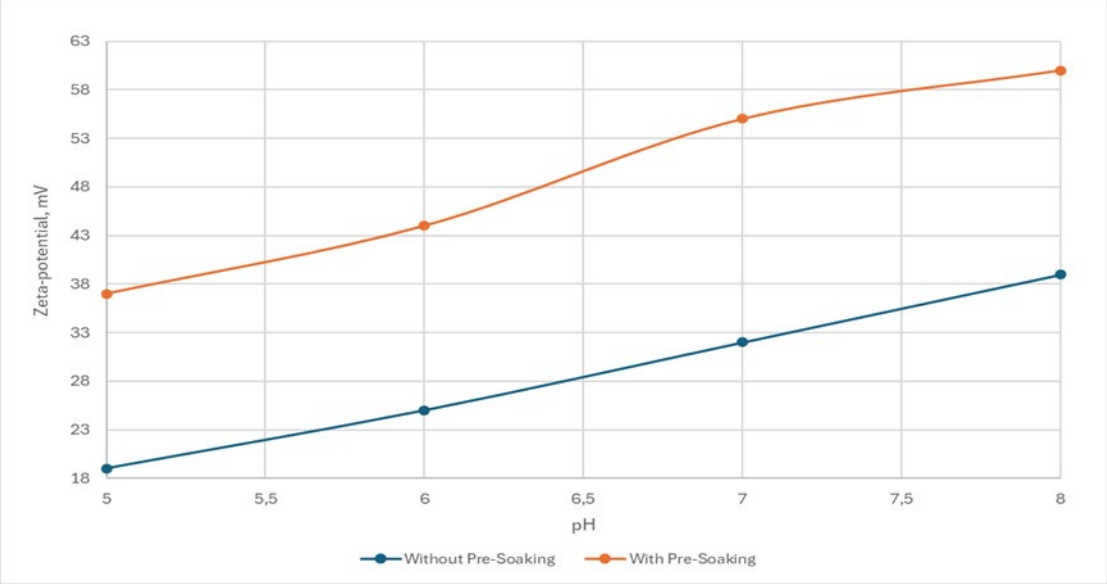


Figure 8. Experimental Zeta-potential data for crushed coal, pH = 5–8.

As shown in Tables 4 and 5, and Figures 7 and 8, the secondary Rehbinder effect in the studied coal is reflected by a shift in the hydrophilic-hydrophobic balance of the total external surface of the crushed coal toward greater hydrophilicity. This is likely caused by the exposure of oxidised pore surfaces through the primary Rehbinder effect, which, following the crushing of pre-soaked coal, contribute their oxidised fraction to the external surface.

The experimental results indicate that a stable, highly concentrated water-coal slurry (water-coal fuel) can be prepared from low-metamorphosed Donetsk coal at pH > 4 (ζ > 30 mV) when the coal is pre-soaked and at pH > 6.7 when dry-crushed.

4. Conclusions

The experiments have confirmed that the Rehbinder effect productively weakens the organic matter of low-metamorphosed coal from Donetsk basin. However, a quantitative evaluation and prediction of organic matter comminution across different coal metamorphism grades would require expanding the experimental scope to account for several factors:

First, the characteristics of pores and fractures in the coal matter, which are exposed by liquid films, as well as the parameters of these pores depend on the degree of coalification. Second, it must not be overlooked that infiltration of liquid (water+reagents) into coal pores and fractures is influenced by the hydrophilic-hydrophobic properties of the pore surfaces, as well as by the composition and properties of the infiltrating liquid and any reagents used. Third, the kinetics of liquid infiltration into coal pores and fractures must be considered, as this allows to determine the required duration of effective coal soaking (during which the weakening effect of the Rehbinder mechanism is significant).

The experiments have demonstrated that the secondary Rehbinder effect in the studied coal is expressed as a shift in the hydrophilic-hydrophobic balance of the total external surface of crushed coal toward increased hydrophilicity. This is likely caused by the exposure of oxidised pore surfaces through the primary Rehbinder effect, which, following the crushing of pre-soaked coal, contribute their oxidised fraction to the external surface.

Analysis of the experimental data shows that a stable, highly concentrated water-coal slurry (water-coal fuel) can be prepared from low-metamorphosed coal from Donetsk basin at pH > 4 (ζ > 30 mV) when the coal is pre-soaked and at pH > 6.7 when dry-crushed.

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