Original Article

Permeation distance: complementary methodology for assessing the thermoplasticity of metallurgical coals

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Abstract

The present paper aims to present a new methodology to measure the thermoplasticity of coals by the permeation distance method. This development is a reproduction of a methodology created at JFE Steel and adapted to the reality of the coals used for the production of metallurgical coke in Brazil. This method consists of evaluating the thermoplastic behaviour of coals in a complementary way to Plastometry and Dilatometry analyses. Experiments were carried out for coals of different ranks and petroleum coke. Furthermore, the hypotheses of additivity and tests with different particle sizes for the same coal were validated. It was concluded that there are significant differences between permeation distances for coals of the same fluidity. It is necessary to bring emphasis for the anomalous behaviour of the petroleum coke (CVP). The granulometry is a relevant factor for the permeation distance with a direct impact on the quality of the semi-coke, with a direct influence on the mechanical strength of metallurgical coke.

Keywords: Permeation distance; Plastometry; Coke quality; Reology.

1 Introduction

Within the context of steelmaking in the world, the steel production route using blast furnaces is still predominantly used, the use of coke as the main raw material as a source of carbon to produce hot metal means that research with a commitment to improving coke quality is increasingly necessary. To do this, it is necessary to use coal, which is the raw material for coke production, and use a specific coal with coking properties, that is, it must solidify again after the plastic phase during the coking process. The difficulty of finding all the necessary characteristics in a single coal that allows the production of coke with the quality required by the blast furnace means that, in practice, coal blends are necessary formed with two or more coals [1]. It is possible to catalog coals according to their coking capacity. They are divided into: hard coking coal, semi-hard coking coal, soft coking coal, semi-soft coking coal, and PCI coal [2].

The variability of each type of coal is reflected in the set of its physical, chemical and petrographic characteristics, resulting in a composition of blends where there are different types of coal.

The measurement of coal thermoplasticity using the permeation distance methodology was developed with the aim of simulating the plastic behavior of coal under conditions close to those observed during the metallurgical semi coke formation process.

This work is a reproduction of the methodology created by the JFE Steel Research and Development Center in Japan. A technical knowledge transfer program signed between JFE and Gerdau – Unit of Ouro Branco supported the reproduction of the methodology. The main objective is to analyze the plastic behavior of coal by observing its ability to fill the empty spaces between glass spheres that simulate adjacent particles of inert coal and, ultimately, measure the permeated distance after the sample has cooled. This test is recommended for the evaluation of high-volatile coals that have high fluidity as a complement to the evaluation of rheological aspects, essential for defining blends aimed at obtaining coke of suitable quality for use in blast furnaces. Fluidity is a measure of the binding ability that involves the coal interface area between particles and is a function of the coal rank, the relative amounts of reactive and inert macerals and by the heating rate, constant in the case of the plastometer [3].

One of the issues with the Gieseler Plastometry test is the low viscosity of high-fluidity coals, especially above 40,000 ddpm, behaving like a pseudoplastic material, resulting in the Weissenberg effect (Figure 1), where the stirring rod rotates so fast that it leads to the formation of a vortex in the analyzed fluid [4]. The rod rotates in such a way that there is no fluid in contact with it, affecting the reliability of the fluidity test result [4].

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In this context, Permeation Distance emerges with the proposal to measure the thermoplasticity of coal by simulating the conditions around thermoplastic coal during the coking process through the interaction of coal with a layer of glass spheres with the same average particle size as the coal sample [4], as shown in Figure 2.

For the experiments carried out and presented in this paper, we replicated the methodology developed by JFE. The results obtained in studies by Dohy et al. [4] from 2014 and Dohy et al. [5] from 2020 stated that coals with high fluidity had a very large permeation deviation. Furthermore, high fluidity (MF) coals with a greater maximum permeation distance form thinner pore wall structures in the coke, and the strength of the coke deteriorates when the coal blend includes coal with a greater maximum permeation distance.

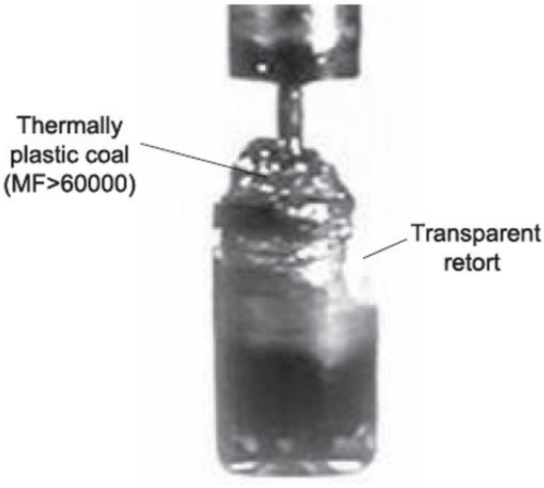


Figure 1. Weissenberg Effect in the retort during the Plastometry test. Reproduced from Dohy et al. [4].

To add relevant information to this paper, different types of coals with different fluidity values were tested, using low volatile, medium volatile and high volatile coals. A product derived from petroleum, used in coke ovens, was also tested: petroleum coke. As it is an important component in the coal mixture in some cases, there was a need to test it to collect information that could be crucial to producing quality coke.

In order to obtain more information, different coal particle sizes were tested.

In addition to additivity tests with two coals of different fluidity to analyze the influence of the permeation distance during the procedure.

2 Materials and methods

2.1 Permeation distance methodologies

The apparatus consists primarily of an electric furnace for heating the coal sample, moreover, permeation distance measurement device. The main monitor integrated into the electric furnace provides control over the furnace temperature, nitrogen flow rate. In addition, the information such as temperature elevation rate, sample identification, data editing and export commands to a computer via a USB connection [4]. Additionally, this equipment contains a

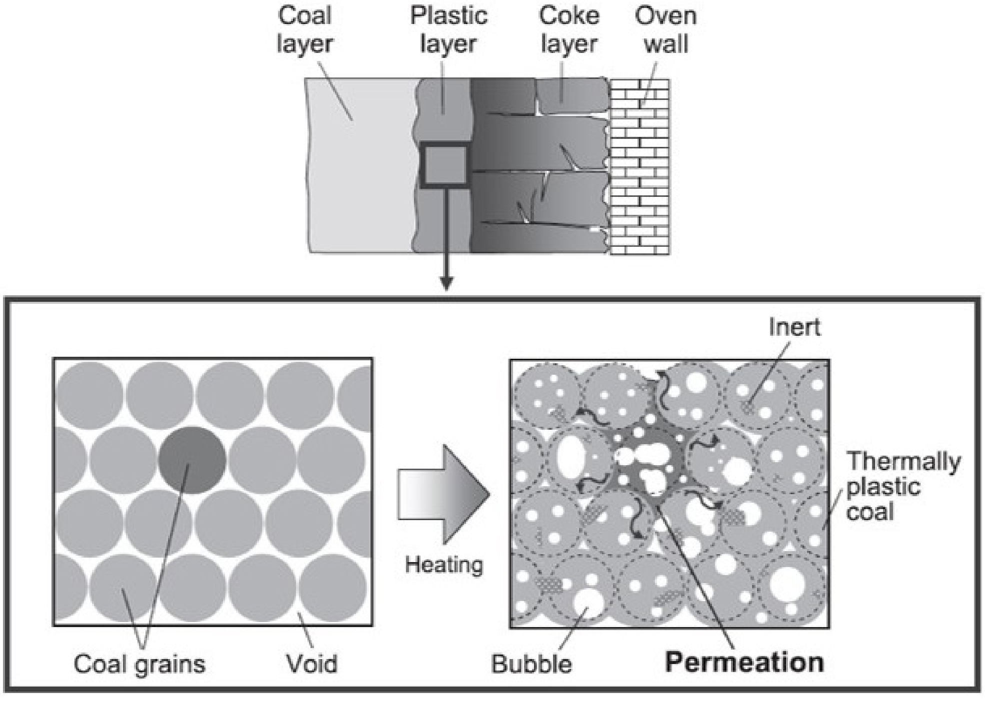


Figure 2. Conceptual diagram of coal thermoplasticity in coke oven. Reproduced from Dohy et al. [4].

camera for internal monitoring of the sample during the test, allowing real-time observation of permeation behavior during the test [4]. This equipment also has a piston that attaches to the sample and exerts a force of 1.6 kgf on it, simulating the load pressure in a conventional furnace. Figure 3 illustrates the constructed permeation distance equipment [4].

The experimental procedures are described as follows [4]:

- 2.5 grams of crushed and sieved coal sample (particle size < 2mm) are used. This mass is loaded into a quartz tube with a diameter of 20mm and a height of 100mm. The sample is compacted to reach a layer thickness of about 10mm;
- Glass spheres with a diameter of 2mm are weighed using a precision balance to obtain 25g with a variation of 0.05g. They are then placed above the packed layer to achieve a thickness between 54mm and 57mm;
- A quartz filter with a diameter of 20mm and a thickness of 5mm is placed above the glass layer, and a quartz rod is placed on the filter, exerting a force of 1.6 kgf on the assembly.

Figure 4 provides a schematic representation of the tube used and a real image.

Subsequently, the test tube is placed in the middle of the electric furnace. After that, it is necessary close the door and check all the security locks. After this, the oven heating process must begin by turning it on to the oven's digital display. Adjust the oven to heat up to 600 °C at a heating rate of 3 °C/min under a nitrogen atmosphere. During heating, the coal in the plastic phase permeates the glass sphere layer. After the sample has cooled, the permeated distance is measured using a graduated ruler in millimeters. The semi coke formed between the glass spheres is not recovered after cooling, but the semi coke formed at the bottom of the quartz tube, within the initial 10mm range of coal, is partially recovered.

The weight of the glass spheres after the test directly affects the equation used to determine the permeation distance. The permeation of the recovered mass is defined according to Equation 1.

$$\boldsymbol{D} = \boldsymbol{H}\left(\boldsymbol{G} - \boldsymbol{M}\right) \tag{1}$$

where: D is the maximum permeation distance (mm); H is the height of the packed layer per 1g of packed glass spheres in this experimental crucible (mm/g); G is the weight of the initially packed glass granules (g); M is the weight of the granules that did not adhere along with the semicoke (g).

Below is an illustrative example of the test before and after permeation (Figure 5).

2.2 Materials and evaluated blends

For the tests conducted, it is used low, medium, and high-volatile coals, as well as petroleum coke in two particle size distributions, below 2mm and below 1mm, with the aim of understand the influence of particle size on permeation distance. In addition to individual material tests, it was tested blends to evaluate the interaction between coals and petroleum coke and test the hypothesis of permeation distance additivity. Table 1 presents the materials used in this study, the Table 2 presents the samples used and particle size, and Table 3 presents the blends for additivities tests.

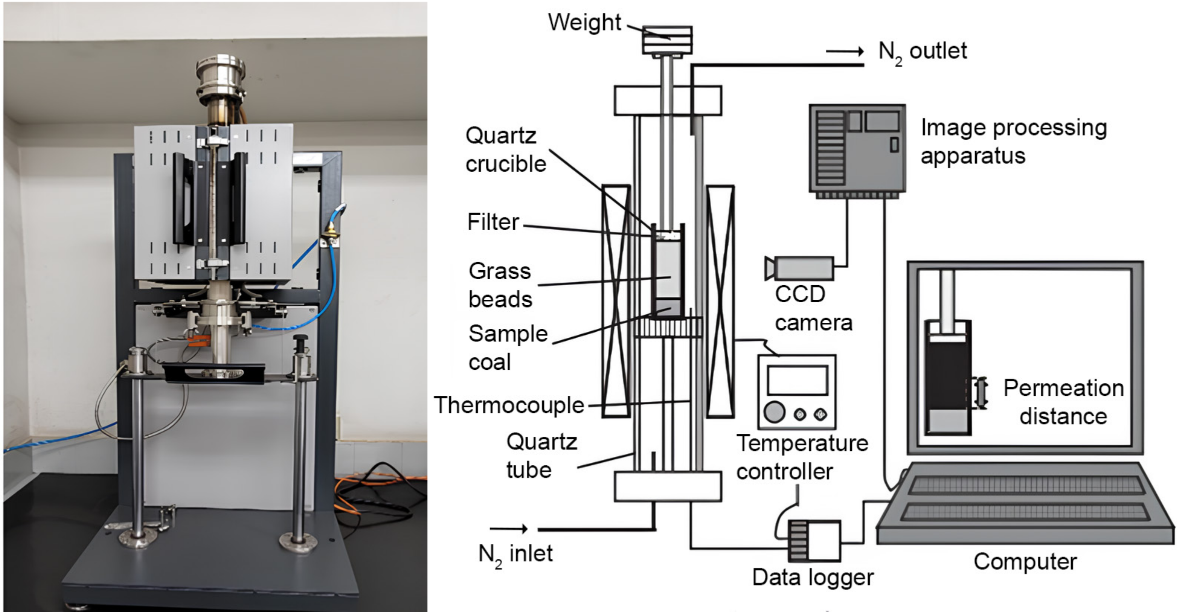


Figure 3. Permeation distance measurement apparatus reproduced in Brazil and schematic model of the permeation distance equipment. Reproduced from Dohy et al. [4].

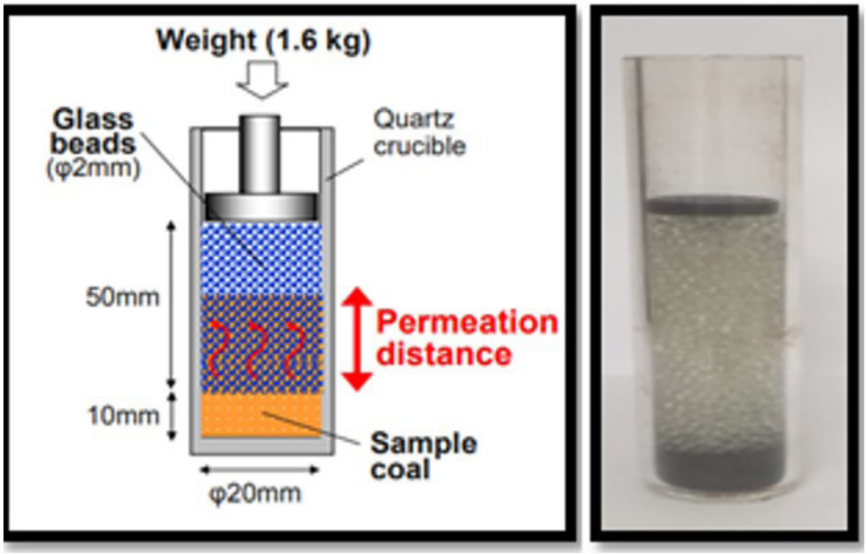
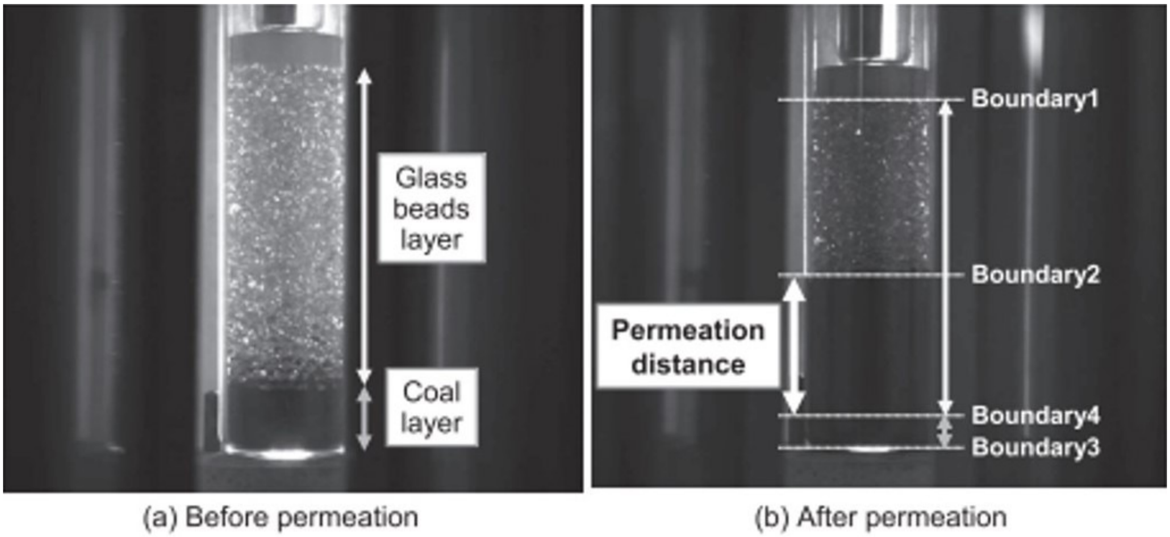


Figure 4. Quartz tube before the permeation distance test. Reproduced from Dohy et al. [4].



(a) Before permeation

(b) After permeation

Figure 5. Schematic illustration of coal permeation. Reproduced from Dohy et al. [4].

Table 1. Coals used in the permeation distance tests

Coal	VM (%)	Ash (%)	S (%)	logMF (logddpm)
AV-1	35.85	8.28	1.90	4.56
AV-2	31.38	7.45	0.94	4.30
AV-3	33.32	6.76	0.78	4.14
AV-4	24.00	9.39	0.35	4.44
MV-1	22.50	10.50	0.64	2.70
MV-2	24.34	9.49	0.85	2.53
BV-1	20.46	9.96	0.68	2.70
CVP	12.60	0.895	0.83	0

VM: volatile matter; S: Sulfur; logMF: log maximum fluidity.

3 Results and discussions

The papers developed by Dohi et al. [4,5] showed that there was a wide variation in permeation distances according to the materials analyzed, especially with high volatile coals. For this paper, the same phenomenon observed for a type of high volatile coal, other high volatile coals remained in a close range, and with greater permeation distance values in relation to medium volatile and high volatile coals. This confirms a conclusion from previous papers stating that coals with greater fluidity have a greater permeation distance.

Analyzing other key points for understanding the permeation distance was testing the particle size of the sample where even materials with different particle sizes compared and the results indicated were a shorter permeation distance with a smaller particle size, which explains a better filling of the pores forming a more resistant semi-coke.

Table 2. Samples used in the tests and the granulometry used

Samples	Coals	Particle size
1	AV-1	100% < 2mm
2	AV-2	
3	AV-3	
4	AV-4	
5	MV-1	
6	MV-2	
7	BV-1	
8	CVP	
9	CVP	100% < 1mm
10	AV-1	
11	AV-2	
12	AV-4	
13	BV-1	

Finally, tests analyzed with samples composed of two different coals to analyze the influence of a mixture. For coals with different volatilities, the result was very close to an average of their separate permeation distances, which shows consistency between the results found.

These results found were consistent with the papers by Dohy et al. [4] from 2014 and Dohy et al. [5] from 2021.

For tests with petroleum coke, known in Brazil as CVP, and used in some coal mixtures, there was an opposite effect to other coals.

There was no permeation but rather a contraction effect. In the mixture test, in a sample of coal and CVP, the contraction effect of the CVP observed, where there was no permeation.

The results of the measurements on the millimeter scale identified in Table 4; each sample is respectively demonstrated according to the distance obtained after the test more over. Figure 6, the results for permeation distances for the tested coals the blue dots represent the coals and the red dot represents the CVP, drawing attention to its contraction during the experiment. Figure 7, we have a schematic representation of the presumed behavior of a high-fluidity coal in its thermoplastic state for distinct maximum permeation distances, which reflect in the formation of the coke microstructure [4].

Table 3. Blends for additivities tests

Blends	BV-1	AV-4	AV-1	CVP
BV-1 + AV-4	50%	50%	-	-
BV-1 + AV-1	50%	-	50%	-
CVP + BV-1	50%	-	-	50%

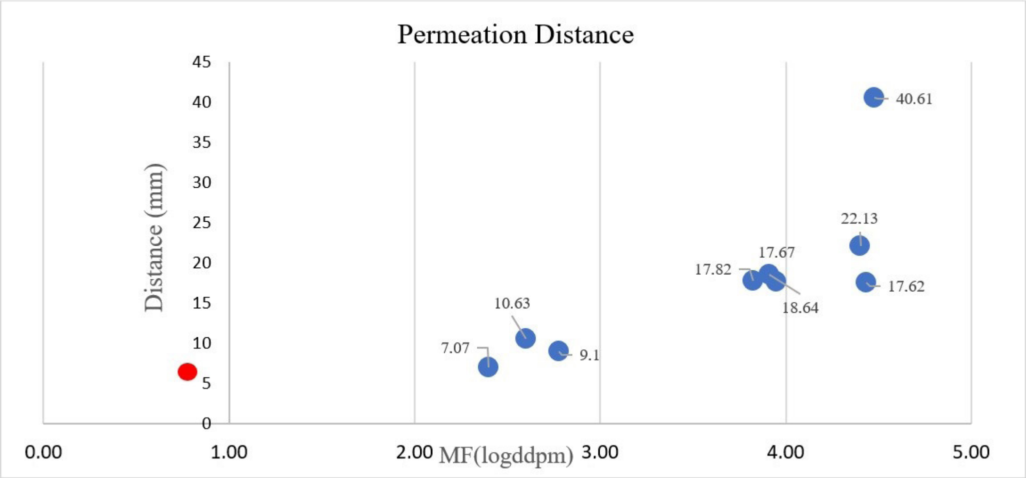


Figure 6. Permeation Distance chart.

In the case of long maximum coal permeation distances, there is a greater spread of material mass during devolatilization/swelling, leading to an increase in the average pore size of the coke, which tends to make its microstructure more fragile [1]. On the other hand, for coal with the same fluidity but a shorter permeation distance, the pore size distribution is more suitable, benefiting the coke's strength. Thus, permeation distance can complement classical rheological analyses in evaluating the Value in Use (VIU) of each coal. For CVP, an anomalous behavior observed, contrary to permeation, showing contraction at the end of the test the CVP presents this contraction because it has a different structure than coal, a more porous structure where the coal coalesces between these pores and thus forms a semi-coke and a more compact coke at the end of the process.

Regarding the additivity tests, Figure 8 presents the results for binary blends (50/50%) of coals. In a Figure 9 presents the result for blend with coal and CVP. For coal blends, the permeation distance is very close to the average of the individual coal distances.

Table 4. Results of permeation distance

Coal	Distance (%)
AV-1	22.13
AV-2	40.61
AV-3	17.67
AV-3	18.64
AV-4	18.64
AV-5	17.82
AV-6	17.62
BV-1	9.10
MV-1	10.63
MV2	7.07
CVP	-1.00

For blends with CVP, a negative deviation from additivity observed, demonstrating that the contraction of petroleum coke significantly influences the permeation of the blend.

Figure 10 shows the variation in permeation distances for coals with two particle size levels, with a top size of 2mm (standard) and 1mm.

It observed that the permeation distance decreases with the reduction in the average particle size due to the lower swelling capacity and resistance potential to internal pressure related to volatile matter release during the plastic regime [5]. In other words, for coals with higher fluidity, it is interesting to increase the degree of comminution to reduce the permeation distance with a focus on forming microstructures more suitable for coke quality [5]. Figure 11 illustrates these two opposing behaviors.

When heated, the coal blend becomes thermally plastic at around 400-500 °C and generates pores in the coke's microstructure [1,2].

Particles of coal with long permeation distances excessively permeate the voids around them. Simultaneously, the pores coalesce and form larger pores, considered defects – points of fragility resulting in a decrease in coke's mechanical strength [5]. In Figure 10b, with lower permeation and initial mass spread, the intensity of pore coalescence is lower, favoring an increase in coke strength [5].

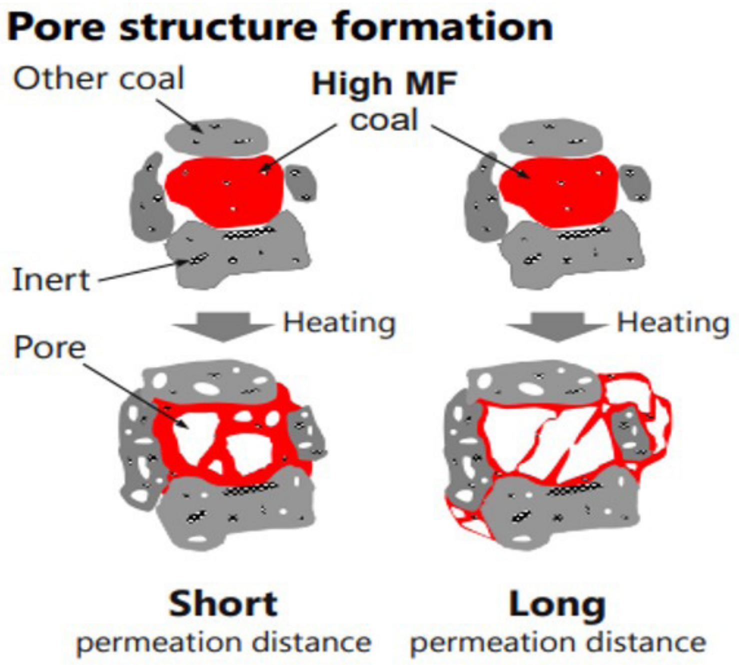


Figure 7. Schematic illustration of behavior of long and short maximum permeation distance coal. Reproduced from Dohy et al. [4].

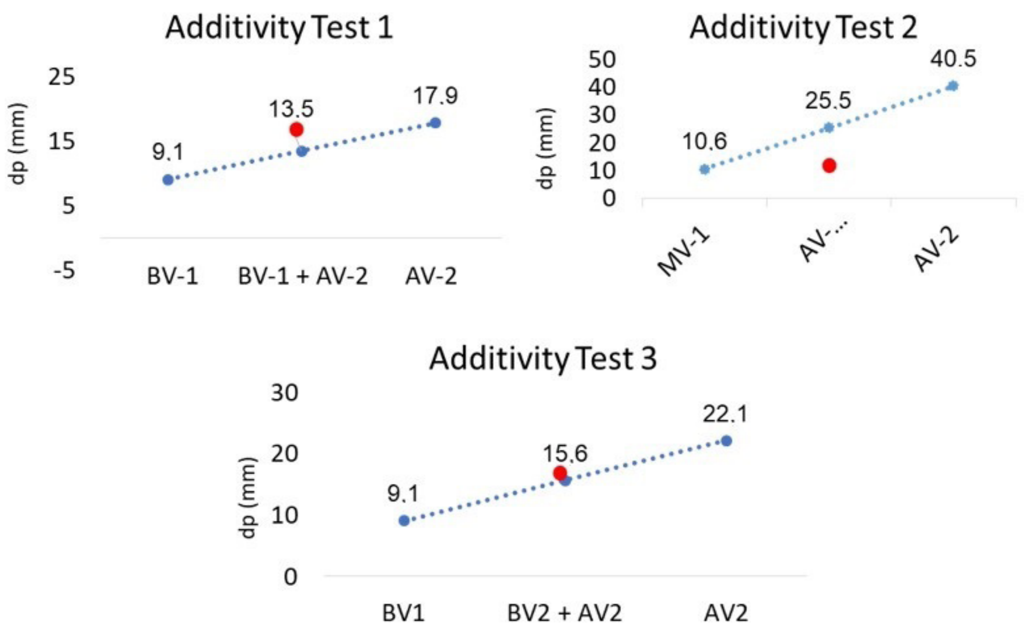


Figure 8. Additivity test for coal blends.

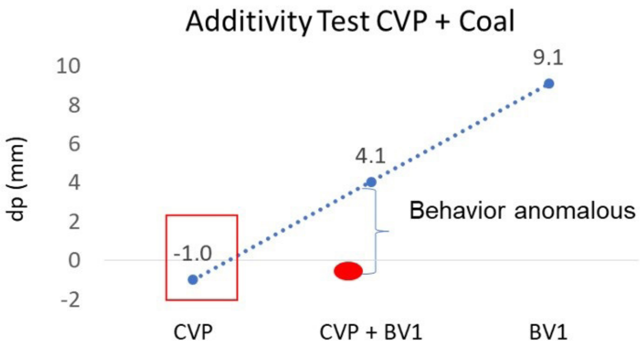


Figure 9. Additivity Test Coal + CVP.

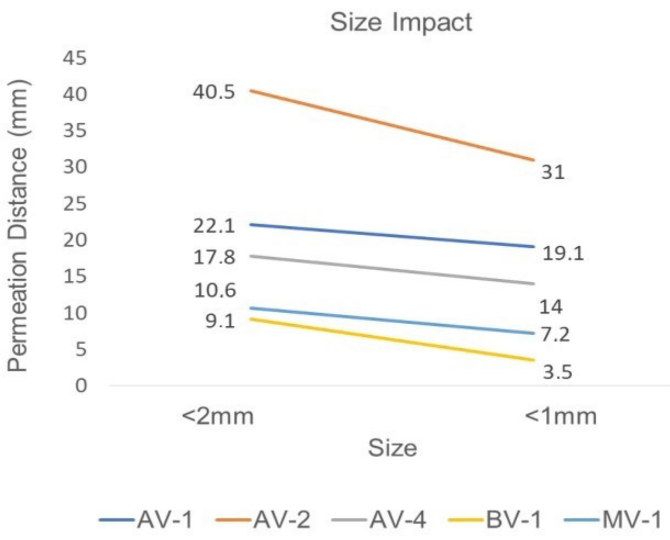


Figure 10. Permeation distance vs. particle size.

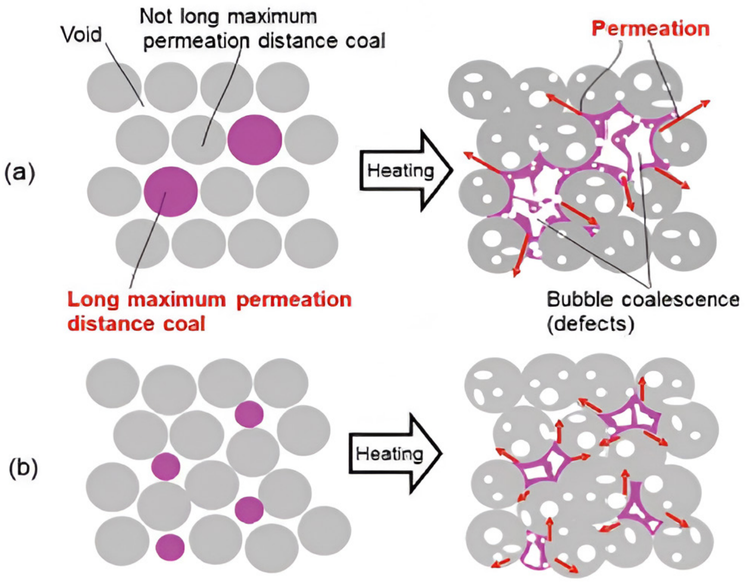


Figure 11. Presumed mechanism of suppression of negative effect of long permeation distance coal on coke strength by reducing grain size of coal blend (a) Mechanism of negative effect, (b) mechanism of suppression of negative effect by reducing grain size of long maximum permeation distance coal Reproduced from Dohy et al. [5].

4 Conclusion

- The permeation distance method proved to be efficient, serving as a complementary analysis to measure the thermoplastic behavior of coals, especially high-volatile ones;
- The application of the "permeation distance" contributes to a more precise assessment of the coal's potential to increase the mechanical strength of coke;
- Coal with long permeation distances increases the porosity of coke, directly influencing the material's mechanical strength;
- Permeation distance is an additive property for coals;
- The use of CVP favors the contraction of the semi coke, demonstrating a negative deviation from additivity;
- The maximum permeation distance is reduced with the decrease in coal particle size, promoting the formation of a more refined microstructure suitable for mechanical coke quality.

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