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EFFECT OF FLOTATION PROCEDURE AND COMPOSITION OF REAGENTS ON YIELD OF A DIFFICULT-TO-FLOAT COAL

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The paper discusses two procedures of flotation of difficult-to-float oxidized coal. The first one is the normal flotation procedure which relies on wetting coal with water followed by addition of flotation reagents (dodecane, dodecyl tetraoxyethelene ether ($C_{12}E_4$), and 1-pentanol). The second, direct contact procedure consists of mixing pure reagents with dry coal followed by addition of water. Investigation showed that for both procedures and applied chemicals, the yield – reagent dosage curves reached a plateau. The yield plateau level was 10 g/kg for normal flotation and 20 g/kg for direct contact flotation. At the plateau dosage, normal flotation provided a maximum clean coal yield of only 70% with mixed two-reagent ($C_{12}E_4$ + 1-pentanol). The second procedure resulted in a maximum clean coal yield of ~94 % using the same two reagent but at a higher dosage. It was also concluded that always two-reagent systems provided better flotation yields compared to one-reagent and three-reagent systems. Thus, flotation of difficult-to-float oxidized coal can be successfully accomplished by applying the direct contact flotation procedure with appropriate reagents.

Key wards: flotation, flotation reagents, difficult-to-float materials, oxidized coal

INTRODUCTION

Flotation is one of the most effective techniques for upgrading coals (Davis, 1948; Aplan, 1977 and 1997; Vamvuka and Agridiotis, 2001). For low rank and oxidized coals, however, the process requires certain improvement (Chander, et al, 1994). Various modifications have been proposed to increase flotation of difficult-to-float oxidized coals. The most important include the use mixtures of reagents (collector – co-collector, collector – promoter and collector – promoter-surfactant systems) and two-stage addition of emulsified oily-collector with frother (Firth et al, 1979; Fuerstenau, 1981; Majka-Myrcha and Sobieraj, 1987; Moxon and Keast-Jones, 1986;

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Diao and Fuerstenau, 1992; Aktas and Woodburn, 1994). The use of certain reagent mixtures (oils + ionic collectors or oils + non-ionic surfactants) has been well-documented in literature (Moxon et al. 1988; Chander et al. 1996; Vamvuka and Agridiotis, 2001; Murat et al., 2003). The success of such mixtures to achieve acceptable results was explained by co-adsorption with formation of a mixed film at the tested material surface, which improves adsorption density and increases hydrophobicity (Rao and Frossberg, 1997; Jia, et al., 2000).

Studies related to modification of flotation procedure to improve coal flotation are rare (Mohanty et al. 1998). For this reason, the aim of this work is to improve flotation of highly oxidized coals by modification of the way the reagents are contacted with coal coupled with changing the composition of the chemical reagents used in flotation.

EXPERIMENTAL

REAGENTS

Three categories of reagents were used for flotation of coal: hydrocarbon (dodecane), alcohol (1-pentanol), and dodecyl tetraoxyethelene ether ($C_{12}E_4$). The hydrocarbon usually serves as the collector, alcohol as frother while C_xE_y as modifier. They all were of commercial grades and were used without further purification. The properties of the used reagents are listed in Table 1. Stock solutions of reagents used in the experiments were prepared fresh everyday.

SOLID MATERIALS

Coal used in this investigation was obtained from the Kazimierz Juliusz Mine in Sosnowiec, Poland. It was an oxidized and difficult-to-float material. The bulk sample approximate chemical analysis reflected 3.5% ash content and 10.8 % moisture. The size distribution of coal used in flotation tests is shown in Table 2.

Table 1. Manufacturer properties of the different used reagents at 25°C

Reagent	Producer	M.W. (g/mol)	Phase	Solubility in water	Density g/cm^3
Dodecane ($C_{12}H_{26}$)	Fluka	170.34	Colorless liquid (boils at 216 °C)	Insoluble	0.749
$C_{12}E_4$ ($C_{20}H_{42}O_5$)	Fluka	362.23	Viscous liquid (viscosity 35 cP)	Insoluble (self-dispersed acts as emulsifier)	1.109
1-pentanol ($C_5H_{12}O$)	Fluka	88.15	Colorless liquid (boils at 137.3 °C)	Moderately soluble (22.66 g/dm^3)	0.811

Table 2. Size distribution and ash analysis of the investigated flotation feed

Size, mm	Wt. %	Ash %
-0.500+0.250	13.69	1.56
-0.250+0.150	15.81	2.36
-0.150+0.106	12.78	2.52
-0.106+0.075	17.87	3.62
-0.075	39.85	4.50
Total	100.0	3.35

METHODS OF FLOTATION

Flotation tests were carried out in a mechanical sub-aeration laboratory flotation machine equipped with a 1-dm^3 capacity cell, fed with 100 g dry coal resulting in a solid / liquid ratio of 10 % during flotation. After determining the intended operating conditions, the test was run according to two different procedures called here normal and direct contact flotation, respectively. At the test end, clean coal concentrate and residual tailings were filtered, dried at 90-100 ^{o}C, and weighed.

The normal flotation was run by the standard well-known flotation procedure, in which coal surface was cleaned first by agitation at high solid percentage (65%) in water for three min. Flotation reagents were used as one, two and three-reagent systems. Achieving the required reagents dosage was accomplished by addition of the needed dosage of hydrocarbon (dodecane), $C_{12}E_4$ (nonionic surfactant), and finally alcohol (1-pentanol). The required dosage of each reagent, calculated on the bases of grams of used reagent per kg of dry coal, was taken from 1 % (wt.) stock solution or emulsion in double distilled water, followed by 3 min conditioning after each reagent addition. Flotation pH was natural. Flotation time was 10 min.

In the second procedure, called here the direct contact flotation, pure reagents were mixed with the dry coal sample manually. Sequence of reagents addition was started with the intended dosage of $C_{12}E_4$ followed by required dodecane dosage and finally 1-pentanol dosage was added. To ensure complete adsorption of the reagent on the coal surface, mixing was continued until no aggregates of coal with reagents were noticed. It took usually from 1 to 3 min after each reagent addition. The reagent-coated coal was transferred into the flotation cell, diluted with tap water, and conditioned for 0.5 min. Finally, aeration was started for initiation of the flotation. Flotation time was kept constant and was 10 min.

Comparing the results obtained from different series was performed by direct plotting the measured response (yield) against variable under investigation. In case of using three reagents simultaneously, the plot was done using the Gibbs triangle. In this plot, the apex of the triangle represents one reagent flotation, while the outside borders represents different ratios of two-reagent mixtures, and finally the interior of the triangle shows flotation results with three-reagent mixtures (Hussin, 2004).

RESULTS AND DISCUSSION

NORMAL FLOTATION PROCEDURE

Flotation results of a difficult-to-float coal using different doses of one-reagent and two-reagent mixtures approach at selected compositions of the mixture are shown in Fig. 1. It is clear from this series that a single-reagent normal flotation procedure leads to a maximum yield of about 37% at a dosage of 10 g/kg $C_{12}E_4$ in spite of the moderate success of using such kind of nonionic surfactants for flotation of other oxidized coals (Jia et al. 2000). At the same time, increasing the reagent dosage above 10g/kg level did not provide any improvement. This reflects the high oxidation state of the coal surface, and the difficulty of its flotation using one-reagent normal flotation procedure. On the other hand, two-regent systems (dodecane and 1-pentanol, dodecane and $C_{12}E_4$, as well as 1-pentanol and $C_{12}E_4$) at a total dosage of 10 g/kg provided a maximum clean coal yields of ~ 50%, 45 and 61% , respectively. These yields are higher than those obtained with one-reagent. Such an increase is usually attributed to the interactions between reagents leading to their improved adsorptions on the coal surface.

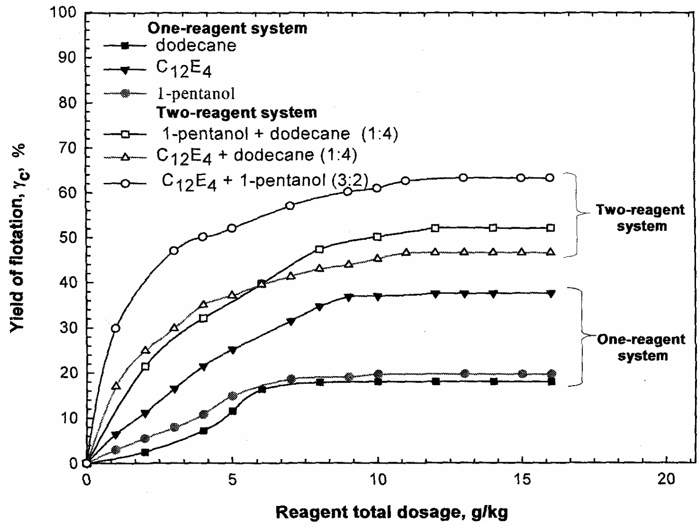


Fig. 1. Effect of one reagent and two-reagent dosage on flotation yield of coal using normal flotation procedure

It can be seen from Fig.1 that in each case there is a maximum yield (plateau), which can be achieved with a given composition of reagents, and the yield is never greater than about 60%. To see whether the yield can be higher in the presence of the three-reagents used in this work, another series of tests was carried out with the oxidized coal at the total dosage of the three-reagent mixtures of 10 g/kg. The results of this series together with the selected data from Fig.1 are presented as the Gibbs triangle in Fig. 2. The plot has the studied reagent combinations marked as circles and the obtained yield values written as numbers beside the circles. From Fig. 2, one can notice that a maximum yield of the clean coal was ~ 61% and was obtained at 6 g/kg $C_{12}E_4$ together with 4 g/kg of 1-pentanol, which is a two-reagent system. Thus, lower clean coal yields were obtained applying one-reagent or three-reagent combinations.

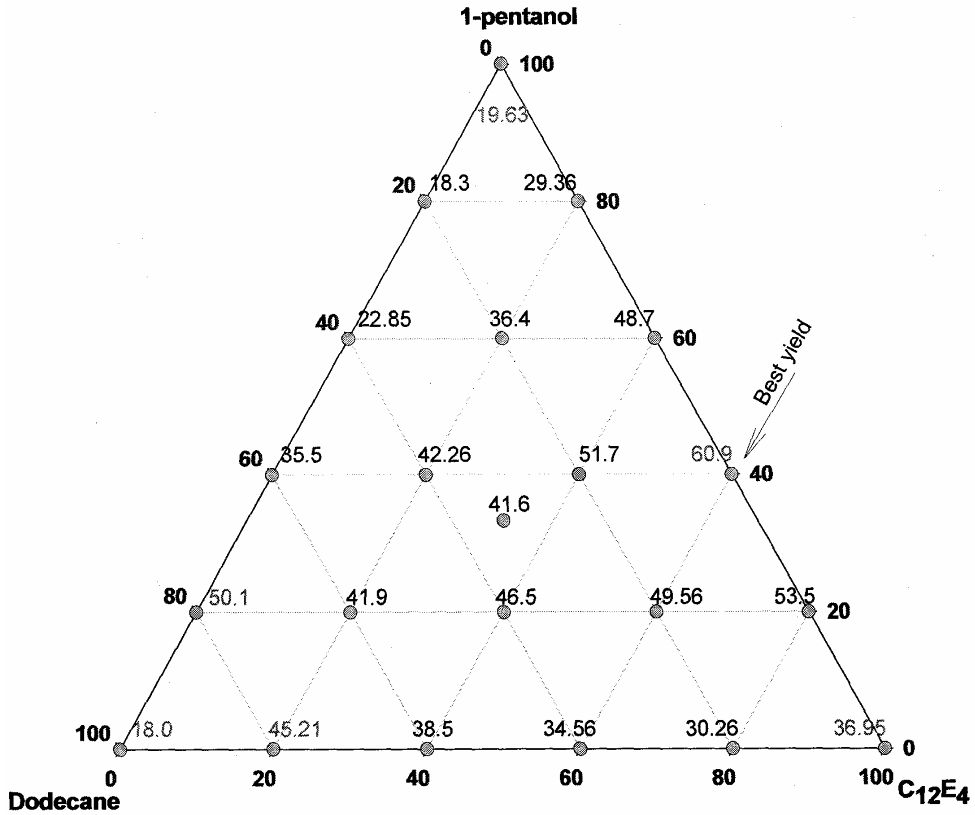


Fig. 2. Effect of dodecane - $C_{12}E_4$ - 1-pentanol combinations on clean coal yield, at a total dosage of 10 g/kg using normal flotation procedure. Ratios among the three reagents are the same as read from ternary plot. Circles represent studied points, numbers show clean coal yield

The relatively poor results obtained applying the normal procedure of flotation with the investigated coal can be attributed to many different factors. The main two parameters seem to be a high oxidation of the coal surface and / or the different not-enough-hydrophobic structures (micelles, emulsions, microemulsions, liquid crystals) formed from the used mixed reagents. Thus, the approach of increasing number of reagents is not successful. Therefore, further investigations were conducted with another procedure of flotation.

DIRECT CONTACT FLOTATION PROCEDURE

Oxidized coals are easy wetted with water and adsorption of hydrophobization reagents is hindered. Therefore we assumed that adsorption of the same chemicals on dry coal surface would be much more efficient and further addition of water to the system would only partially decrease the adsorption. To evaluate the effect of the direct contact procedure in comparison to the normal flotation procedure, we used the same flotation reagents in the tests.

Effect of dosage of one-reagent and two-reagent flotation systems

Figure 3 depicts clean coal yield using different reagents applying the direct contact flotation procedure. The chemicals used were the same as in the normal flotation tests. It can be noticed from Fig. 3 that changing the dosage of one-reagent using direct contact procedure leads to a better flotation yield than applying the normal flotation procedure.

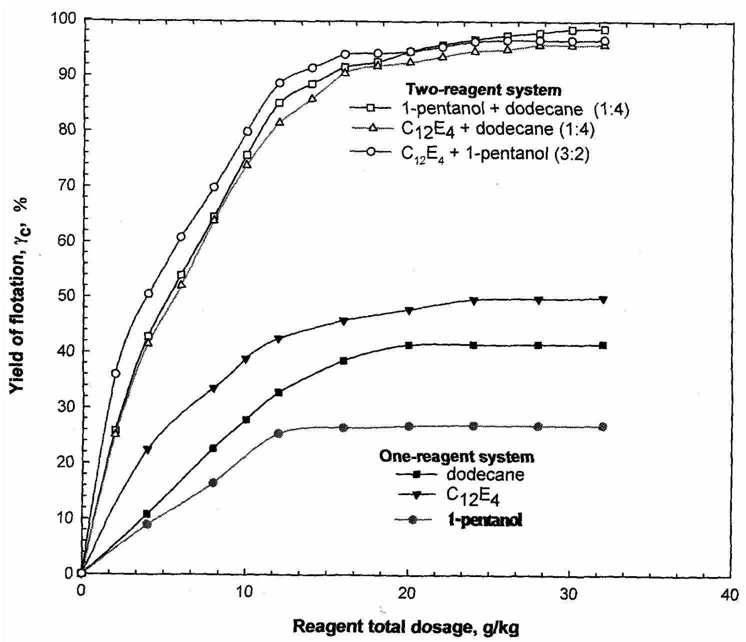
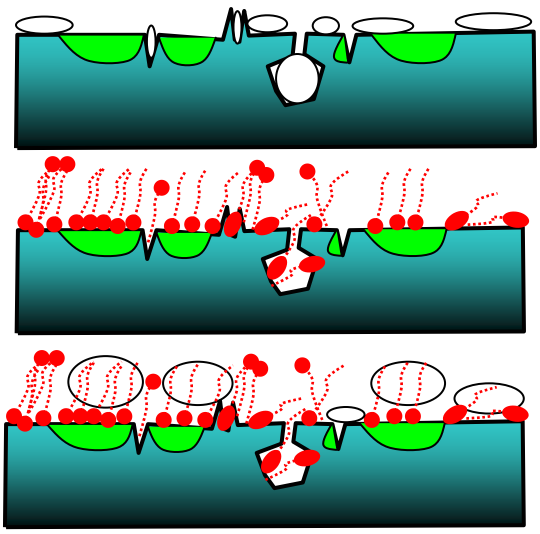


Fig. 3. Effect of one reagent and two-reagent dosage on the flotation yield of coal using direct contact flotation procedure

A maximum flotation yield of ~41.5% at 20 g/kg dodecane was obtained in comparison with a maximum flotation yield of ~18 % at a dose of 10 g/kg when using normal flotation procedure. In the case of nonionic surfactant ($C_{12}E_4$), a maximum yield of ~50% at (20 g/kg) was obtained compared to 37 % at a dosage of 10 g/kg, which also reflects superiority of the direct contact flotation procedure. Reagent interactions in the two-reagent systems investigated in this series reflected a significant positive synergistic effect (Fig. 3). When using dodecane + 1-pentanol, dodecane + $C_{12}E_4$, and 1-pentanol + $C_{12}E_4$ systems, at a total dosage of 20 g/kg, maximum clean coal yields of 94.5, 92.56 and 94.5% were noticed compared to ~ 50%, 45 and 61% for the same systems using the normal flotation procedure. The high yields reflect the negative effect of water during normal flotation of highly oxidized coals. In general, one can draw a conclusion that using single reagent or two-reagent mixtures in the direct-contact flotation procedure leads to an improvement in the clean coal yield but on the expense of reagent dosage used. The high reagent consumption needed by this procedure can be attributed to particles roughness and / or porosity in addition to the thickness of the formed reagent film on the solid surface. It is explained on the proposed adsorption model shown in Fig. 4.



A) Spreading of dodecane droplets take place on hydrophobic sites with possible excess reagents consumption in pores and roughness.

B) Adsorption of $C_{12}E_4$ is possible on hydrophobic and hydrophilic sites with the formation of forms that may hamper flotation and require higher reagent consumption.

C) Adsorption of $C_{12}E_4$ takes place on the oxidized sites giving chance to dodecane droplets to be adsorbed.

Fig. 4. Hypothetical model showing adsorption / spreading of reagents in case of direct contact flotation procedure

dodecane droplets oxidized sites nonionic surfactant

Despite the higher consumption of reagents, when applying the direct contact flotation procedure, it can be considered as successful because it provides greater yields. Further modification of the direct contact flotation for reduction of the reagents is very likely possible. One of the approaches can be the use of a third reagent, as it was done in the previous chapter using the normal flotation method.

Effect of three-reagent mixtures

The presented results showed that there exist two critical dosages of reagents: 10g/kg which represents maximum effective dosage in case of normal flotation, and 20 g/kg in direct contact flotation procedure. For this reason, the same three-reagent combinations used in normal flotation were investigated using direct contact flotation procedure at two different dosage levels, that is at 8 and 16 g/kg, just below the maximum dosage for each procedure.

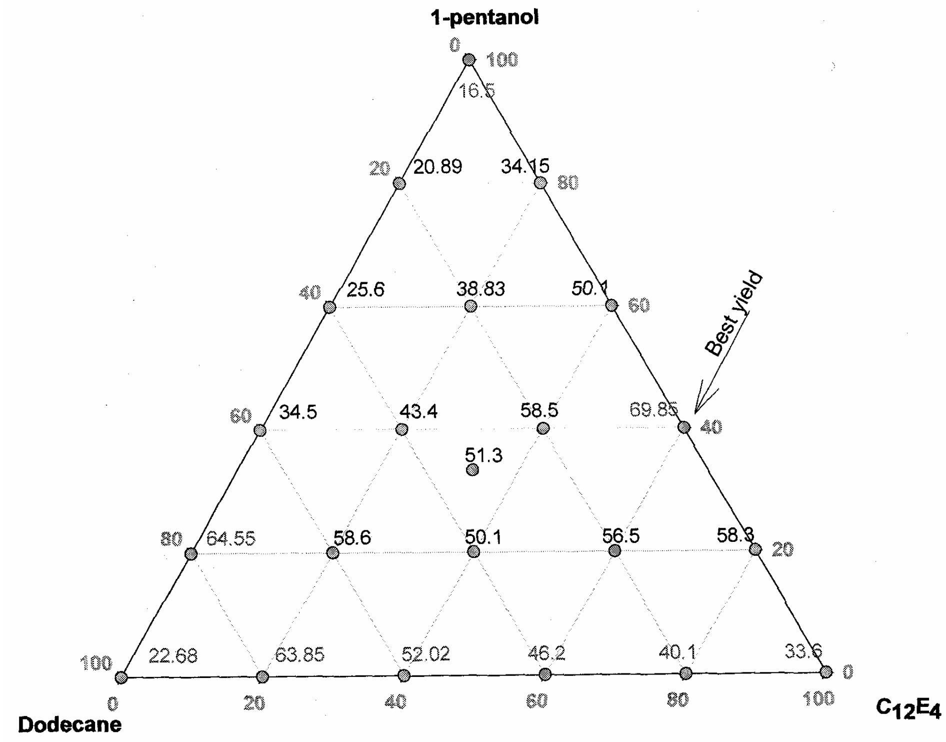


Fig. 5. Effect of dodecane - C $_{12}$E$_4$ - 1-pentanol combinatiats on clean coal yield, at a total dosage of 8 g/kg using direct contact flotation procedure. Ratios (in wt. %) among the three reagents are the same as read from temary plot. Circles represent studied points, numbers show clean coal yield

Figure 5 illustrates the Gibbs triangle plot for the yield obtained from the different combinations at the lower dosage of 8 g/kg. The maximum clean coal yield obtained in both procedures was encountered at the same two-reagent system ($C_{12}E_4$ + 1-pentanol at the ratio of 3:2). The higher maximum yield in case of direct contact (~70%) compared with that obtained in normal flotation (~61%) can be understood taking into account the adsorption model shown in Figure 4. At the same time, all of the studied combinations (using direct contact at 8g/kg) provided higher yields compared to that obtained by normal flotation procedure (at 10 g/kg) except the case of using 1-pentanol only. The lower yield obtained in such a case can be attributed to adsorption of too short chain length of 1-pentanol on coal pores and roughness. Thus, its consumption in coal pores is higher than the other studied reagents. Figure 6 shows yields obtained at the higher dosage level. A maximum yield of 93.85% was obtained at 9.6 g/kg $C_{12}E_4$ together with 6.4 g/kg of 1-pentanol. It confirms the superiority of the $C_{12}E_4$ – 1-pentanol system. It is also clear that the three-reagent combinations did not result in encouraging results even for the direct contact procedure. This is because in all the studied systems and combinations, the highest yield of the cleaned coal was obtained on the lines of the Gibbs plot representing two-reagent combinations.

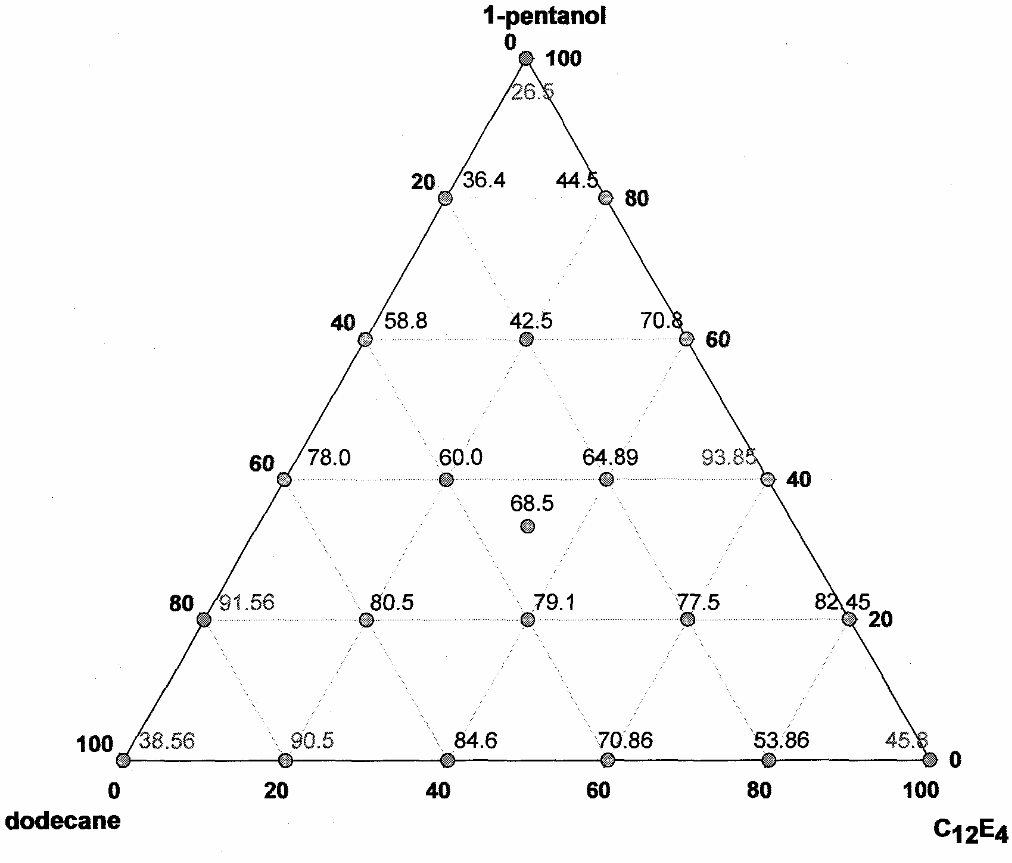


Fig. 6. Effect of dodecane - C $_{12}E_4$ - 1-pentanol combinatiats on clean coal yield, at a total dosage of 16 g/kg using direct contact flotation procedure. Ratios (in wt. %) among the three reagents are the same as read from temary plot. Circles represent studied points, numbers show clean coal yield

CONCLUSIONS

From this study one can draw the following conclusions:

1. One-reagent flotation resulted in poor clean coal yields for both normal and direct contact flotation procedures.
2. After a certain dosage of the reagent under study, 10 g/kg in normal flotation and 20 g/kg in direct contact flotation, the further improvement in clean coal yield can be considered insignificant.
3. Using a two-reagent system improves flotation. Flotation improvement was higher in case of the direct contact flotation.
4. For both of the studied flotation procedures, three-reagent combinations provided relatively poor clean coal yields.
5. The maximum yield of cleaned difficult-to-float coal was ~94% for 9.6 g/kg $C_{12}E_4$ mixed with 6.4 g/kg of 1-pentanol for direct contact flotation, while in normal flotation procedure the maximum clean coal yield was ~ 70%.

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W artykule przedstawiono dwie różne procedury flotacyjne stosowane we flotacji trudno flotowalnego, utlenionego węgla. Pierwsza procedura jest to tradycyjna procedura flotacyjna, która polega na zmieszaniu węgla z wodą, a następnie dodaniu odczynników (dodekan, eter dodecylotetraoksylenu $C_{12}E_4$ i 1-pentanolu. Druga procedura polega na bezpośrednim kontakcie mieszaniny czystych odczynników z suchem węglem, a następnie dodawaniu wody. Badania pokazały, że dla obu zastosowanych procedur, krzywe obrazujące zależność wychodu od ilości dodanego reagentu osiągnęły plateau. Wartość plateau dla normalnej flotacji wynosiła 10 g/kg i 20 g/kg dla flotacji z bezpośrednim kontaktem reagentu. Przy dodatku odczynników odpowiadającym plateau, flotacja prowadzona zgodnie z normalną procedurą dostarcza węgiel z wydajnością 70% w przypadku mieszaniny odczynników ($C_{12}E_4$ + 1-pentanol). Zastosowanie drugiej procedury pozwala uzyskać wydajność czystego węgla ~94% przy zastosowaniu tej samej mieszaniny odczynników, ale w większej ilości. Ustalono, że układy składające się z dwóch odczynników zapewniają lepszy wychód węgla w porównaniu do układów z jednym lub trzema odczynnikami. Flotacja trudno flotowalnego węgla może być z powodzeniem zrealizowana przy użyciu procedury bezpośredniego kontaktu odczynników z węglem.