RHEOLOGY AND STABILITY OF COAL—OIL AND COAL—OIL—ALCOHOL DISPERSIONS

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ABSTRACT

Studies on the effect of various surfactants on the stability of coal—oil mixtures (COM) have been carried out and a few of them were identified as most effective stabilizing agents. Rheological studies on coal dispersions in hexadecane were undertaken as a model system to elucidate the role of these surfactants on COM stability. Viscosity measurements of COM with various percentages of coal revealed that there is a marked increase in viscosity above 40% of coal concentration. At low concentrations of surfactants ($<1\%$) considerable decrease in viscosity of COM was observed. It was concluded that these surfactants are effective in the concentration range 0.1—0.5%, above which the effect levels off. The shear thinning behavior was evident both in the presence and absence of surfactants.

Fuel oil is highly viscous and requires elevated temperatures to exhibit suitable flow characteristics. Methanol or ethanol was found to be immiscible with the fuel oil used in the present study. However, upon the addition of an appropriate amount of a lower chain hydrocarbon or kerosene, a stable single phase mixed fuel system (fuel oil + alcohol + alkane = alcofuel) was formed. The coal + oil mixtures prepared with alcofuels exhibited much lower viscosity than those prepared with fuel oil No. 6. Both types of COMs showed similar heating values. The advantages of alcofuels as compared to those of conventional fuel oils are emphasized in relation to COM technology.

INTRODUCTION

Coal oil mixture (COM) provides a viable and immediate replacement for rapidly depleting petroleum liquids until coal-derived liquids become commercially available. While the technical feasibility of COM as an alternative fuel in utility and blast furnaces has been recognized [1], COM technology often faces the problems of instability and unfavorable rheological properties (e.g., high viscosity problems during processing and pumping). In order to overcome instability, COM technology employs carefully chosen additives. The method essentially involves the screening of several additives (surfactants) in order to impart stability against sedimentation of coal particles in COM. The success often depends on complex interplay of several physicochemical parameters like coal type (surface characteristics — philicity, wettability, mineral matter content, moisture content), oil type (density, viscosity and

chemical constitution of components), structure of additives (surfactants), coal particle size distribution, effective adsorption of surfactants on coal surface and COM preparation conditions. In this type of solid—liquid dispersions, the instability is an inherent problem [2]. Nevertheless, the dwindling supply of fuel oils and their increased price make COM an attractive fuel.

The stability of coal dispersions in oil (COM) requires considerations to different classes of environments such as temperature, pressure, shear, and chemical potential. Stability may be defined as invariance of state properties which leads to static, dynamic and reactive stability according to the nature of the field acting upon the system [3]. Although there could be considerable overlapping of three classes of stability, static and dynamic stability of COM are important in relation to storage and transportation, respectively.

The term "stability" of the dispersion involves aggregative and sedimentative stability. In the colloidal suspensions, two types of stability coexist since Brownian motion can impart stability to individual particles (aggregatively stable). However, as opposed to colloidal dispersions, in the case of coarse particle suspensions such as COM, the practical stability (absence of settling) often requires flocculation as an initial stage of network formation. In the absence of such interactions resulting in a three dimensional network, the powders may settle to form a dense sludge. The mechanism by which surfactants bring about flocculation and impart stability is not established in molecular detail. It was shown that effective stabilizers give a network that is open and can flow readily [3]. Several models of COM stabilization, both at fundamental and technical levels have been discussed recently [3—12]. However, additional work on stabilization and rheology of COM appears to be necessary in order to understand the complex interaction of various physicochemical parameters involved in the solid/liquid dispersion. We have studied coal dispersions in fuel oil No. 6 as well as in hexadecane and measured the rheological properties and stability in the presence of surfactants which were found to be effective after several had been screened. One of the reasons for choosing hexadecane oil is the relatively simpler subsidence technique to study the settling behavior. Since fuel oil No. 6 is not transparent, the subsidence measurement can not be made easily. In subsidence, one observes the descent of a sharp interface that develops in a concentrated suspension with time. We report here our results on viscosity and subsidence behavior of coal—hexadecane mixtures, both in the presence and in the absence of surfactants.

The major thrust of the present work is towards the development of a new class of COM with ethanol blended fuel oil No. 6. The development of such a fuel oil blended COM stems from the following two considerations: (1) fuel oil No. 6 is highly viscous requiring elevated temperature (e.g., 150°F) during processing, storing and pumping. This could be easily eliminated by incorporating ethanol into the fuel oil; (2) the use of ethanol as a fuel oil extender is highly desirable to stretch the fuel oil supply. Moreover, ethanol

can be obtained from renewable sources, e.g., biomass conversion. Similar attempts on blending of petroleum liquids and coal-derived liquids are already in progress [13]. It should be emphasized that fuel oil No. 6 and ethanol are incompatible (immiscible) unlike gasoline + ethanol or diesel + ethanol. In this article, we present the results of our preliminary studies on ethanol blended fuel oil in the presence of coupling agents. Such alcohol blended fuels can be referred to as the "alcofuels". The viscosity and heating values of COM made from fuel oil No. 6 and from alcofuel are compared. Though CFOS fuels (Carbonaceous Fuel-in-Oil Suspensoids) by Keller [14], do contain small amounts of alcohol (5%), there are major differences between alcofuels and CFOS fuels. Keller's work essentially deals with the vapor phase coating of coal particles with ethanol prior to mixing with fuel oil No. 6 [15]. The present approach describes a method of obtaining single phase blends of fuel oil No. 6 and ethanol in the presence of a coupling agent. Moreover, the compatible alcofuel blend could contain up to 25% by weight of ethanol.

EXPERIMENTAL

Materials

Coal samples were from a single batch of finely powdered coal obtained from Ohio. The coal had been pulverized to 80% through 200 mesh and 20% through 325 mesh. The coal powder was used as received. The coal density was determined by volume displacement of reagent grade hexane in a specific gravity bottle and was found to be 1.28 g/cm^3. A sample of low sulfur fuel oil No. 6 obtained from Florida Power and Light Co., Florida was used. Its viscosity at ambient temperature was determined by a Brookfield microviscometer (LVT Model) and found to be 2.2 poise. The density of fuel oil determined by a specific gravity bottle at ambient temperature, was 0.98 g/cm^3. Hexadecane used in the present work was reagent grade (Fisher) having density 0.8 g/cm^3 and viscosity 8 cp. Surfactants were used as received from commercial sources. Ethanol used for fuel oil blending was of commercial grade (Fisher). Kerosene (commercial grade) employed for fuel compatibility study was used without further purification.

Preparation of coal—oil mixtures

COM with hexadecane as oil was prepared by mixing various percentages of coal and oil in sample vials and by shaking the sample vigorously. Surfactants were added to oil before mixing it with coal. In all slurries, the particle size of coal was kept constant. Typical coal percentages in COM vary from 10 to 50% and surfactant concentrations range from 0.05 to 2.5% of COM.

Various COMs with fuel oil No. 6 were prepared by mixing fuel oil and 10 to 25% of coal in a sample vial with constant stirring using a magnetic stirrer. Surfactants were added to the fuel oil before mixing it with coal.

Fuel oil blending and compatibility study

Fuel oil No. 6 and ethanol were not compatible as they rapidly separate upon blending. A premix of n-alkane (hexane, decane) or kerosene and ethanol in a specific ratio was tested for compatibility with fuel oil as follows: At a specific concentration of fuel oil (65%), mixtures of ethanol and kerosene in several ratios were prepared keeping their total percentage at 35% (e.g., 25% ethanol + 10% kerosene, 20% ethanol + 15% kerosene) and added to fuel oil No. 6. The blends were centrifuged for 30 min and examined for compatibility by noting the volume of clear supernatant liquid layer separated (incompatible liquid volume) at the top (V_1) and compatible volume of fuel + alcohol + kerosene (or n-alkanes) at the bottom (V_c) and the volume of the blend in the centrifuge tube (V_t). The percentage incompatible liquid volume (V_{inc}) was calculated as follows:

$$V_{inc}(\%) = V_1/V_t \times 100 \qquad (1)$$

In the present work, we arbitrarily fixed a maximum of 1% V_{inc} as a criterion for compatibility. The value $V_{inc} > 1\%$ was used to define incompatible blends. The limit of 1% V_{inc} was further tested by examining the effect of aging on the blend for several months and noting the separated incompatible liquid volume. The observations were found to be in fairly good agreement with centrifugation results in that there was no further separation from samples exhibiting $V_{inc} < 1\%$ upon aging.

Rheological studies

Viscosities of coal—hexadecane mixtures were determined by Brookfield Cylindrical Viscometer (Model RV). Readings on the dial were taken after 1—2 min of shearing. Viscosity was measured as a function of shear rate. Brookfield Cylindrical Viscometer data were reduced to apparent viscosity (cp) by multiplying the dial reading with the viscometric constant. The viscometric constant was determined by the separate experiments using Newtonian fluids. The shear rate ranges from 6 rpm to 60 rpm which is 6.6 s^{-1} to 66 s^{-1} respectively on absolute shear rate units. The maximum viscosity that could be measured was 100 cp at 6.6 s^{-1}.

The viscosities of fuel oil No. 6 + ethanol blends and COM with the blends range from 60 cp to 2000 cp. In order to cover the wide range of viscosities, we employed a Brookfield Cone and Plate Viscometer which can measure up to 2000 cp at 1.15 s^{-1} (0.3 rpm). The shear rate range was from 0.3 rpm to 60 rpm which is 1.15 s^{-1} to 212 s^{-1} in absolute shear rate units. The apparent viscosity was calculated using the viscometric constant obtained by the separate experiments.

For lower viscosity COMs of coal—hexadecane, the Cone and Plate Visco-

meter did not yield satisfactory results, which could be due to the liquid coming out during higher shear rates and settling faster on the plate during shearing.

Stability tests

In the case of coal—hexadecane mixtures, the settling rates were studied using graduated centrifuge tubes of the same dimensions (15 ml). In these tubes, it was easier to record the descent of a sharp interface that readily developed in the suspension with time. Unlike fuel oil No. 6, which is not transparent, the settling behavior could easily be studied. This was referred to as subsidence behavior, which involves the mass settling [3]. For a comparative study of the effect of surfactants on subsidence behavior, and to establish the order of effectiveness, the dimensions of the settling column were kept constant. Parameters noted are: height of clear liquid layer separated at the top (h_1), black dispersed layer (h_c) at time t and the total height (h_t) of the initial COM column at $t = 0$. A plot of h_1 vs time gives the rate of settling of coal in the COM. These measurements are similar to the subsidence volume measurement by Rowell et al. [12].

Combustion energy of coal—oil mixtures

A comparison of heating values (HV) of COM with fuel oil No. 6 and alcofuels was made by determining the heating values of the samples using Parr Bomb. The heat capacity of the Bomb body was first determined by burning benzoic acid (ΔH combustion = 6.318 kcal/g) at 25 atm of oxygen. Since the data were for a comparative purpose, rigorous analysis of products were not carried out.

RESULTS AND DISCUSSION

Coal—hexadecane dispersions

Figure 1 shows the specific gravity of coal—hexadecane mixtures as a function of coal concentration. The linearity of the plot suggests that there is no appreciable interaction between the coal and hexadecane.

The effect of addition of a surfactant (a petroleum sulfonate TRS 10-410) and coal concentration on the apparent viscosity of coal—oil dispersions is shown in Fig. 2. At a coal concentration of 40—45%, a reduction of about 75% in viscosity is observed in the presence of 0.25% TRS 10-410. The effect of the concentration of various surfactants on the apparent viscosity of a coal—hexadecane mixture (40 : 60) is shown in Fig. 3. For all surfactants, the viscosity of dispersion levels off after a specific concentration of the surfactant. This is presumably related to the adsorption of the surfactant on coal particles. Similar viscosity reducing characteristics of surfactants in col-
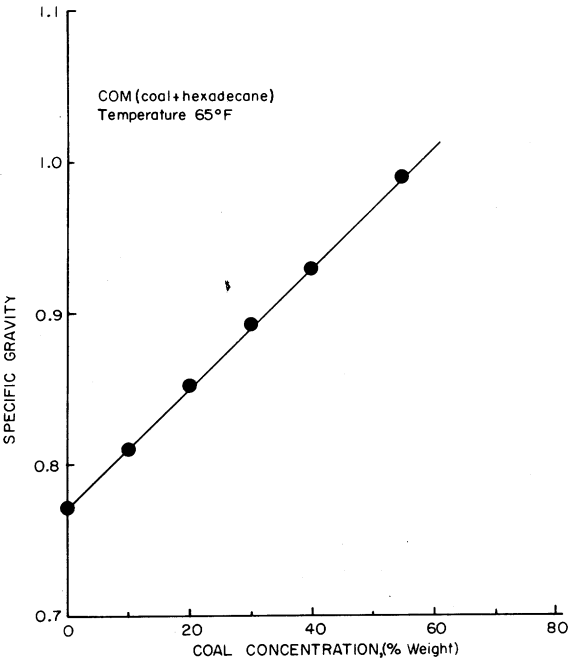
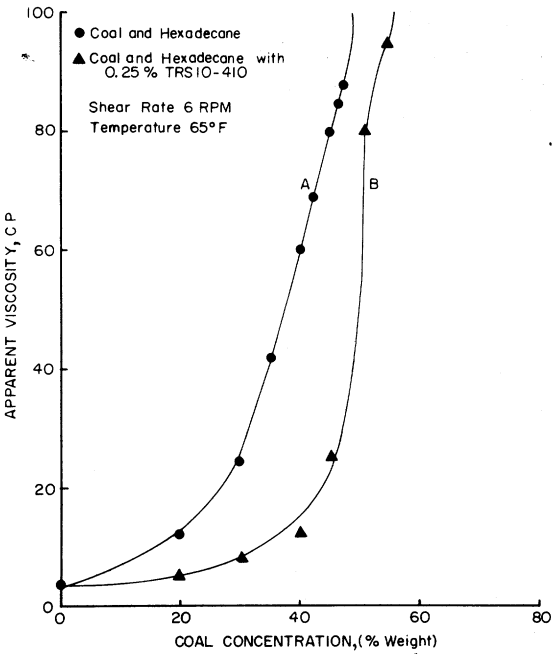Fig. 1. Specific gravity of COM at various coal concentrations.

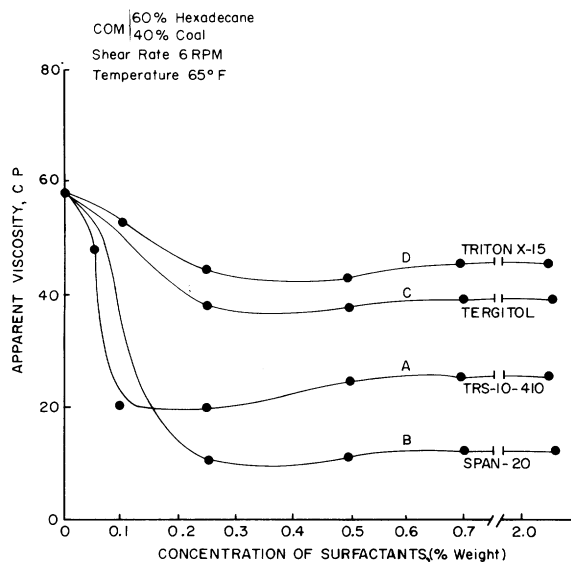Fig. 2. Effect of coal concentration on apparent viscosity of COM.

Fig. 3. Effect of concentration of surfactants on apparent viscosity of COM.

loid suspensions have been reported [16—19]. These investigators have attributed the effect to inseparable electroviscous effects (primary, secondary or tertiary electroviscous effects), invoking the neutralization of the positive or negative charges by surfactant adsorption. As shown in Fig. 4, the coal—hexadecane dispersions exhibited non-Newtonian behavior. The apparent viscosity decreased considerably with increasing shear rates from 6 rpm (6.6 s^{-1}) to 60 rpm (66 s^{-1}). The shear thinning behavior of coal—oil dispersions have been reported by other investigators [4,8,20]. Theoretical aspects and experimental work on rheological properties of suspensions of rigid particles have been reviewed by Jeffrey and Acrivos [21]. Since the data were not sufficient to model the rheological behavior (Pseudoplastic, Bingham plastic, power law fluid, etc.), viscosity dependence on shear rate was considered on normal axes, rather than logarithmic or semilogarithmic axes. Figure 5 illustrates the effect of the ascending and descending shear rate on the viscosity of coal—hexadecane dispersion. The hysteresis observed is similar to the behavior of most thixotropic fluids.

We have examined the settling behavior (subsidence) of coal—hexadecane mixtures in order to compare the rheological and stability characteristics in the presence of various surfactants. Figure 6 shows the subsidence behavior of coal particles in the presence of various surfactants. The petroleum sulfonate TRS 10-410 was found to be the most effective in reducing the subsidence rate of coal in hexadecane. A quantitative comparison of the effect of various surfactants such as TRS 10-410 (anionic), SPAN-20 (nonionic), TRITON X-15 (nonionic) and Tergitol (nonionic) suggests that the most effective surfactant had the lowest supernatant liquid volume and
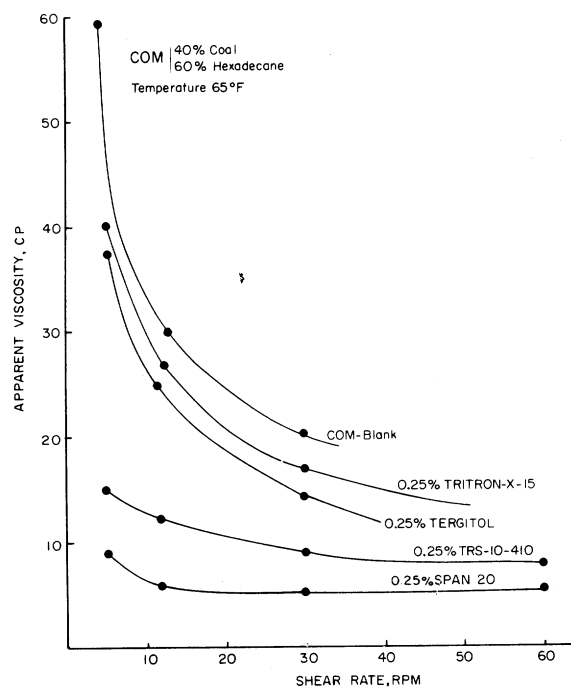
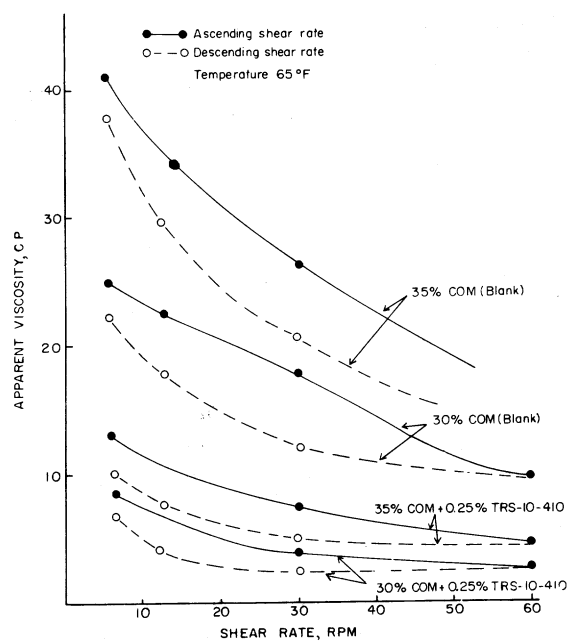Fig. 4. Effect of shear rate on apparent viscosity of COM.

Fig. 5. Effect of ascending and descending shear rate on apparent viscosity.
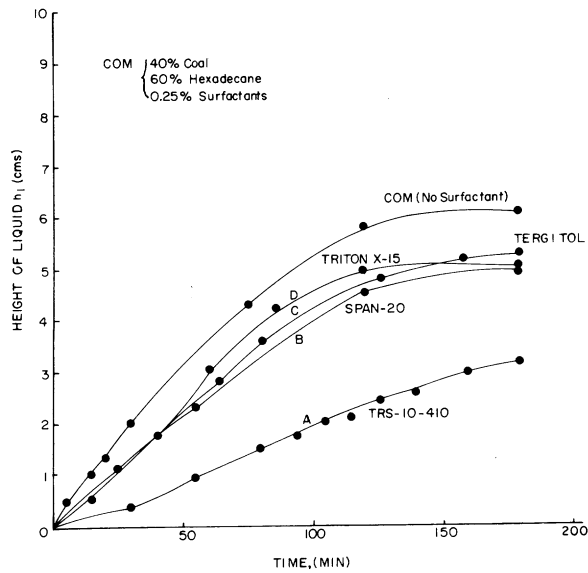
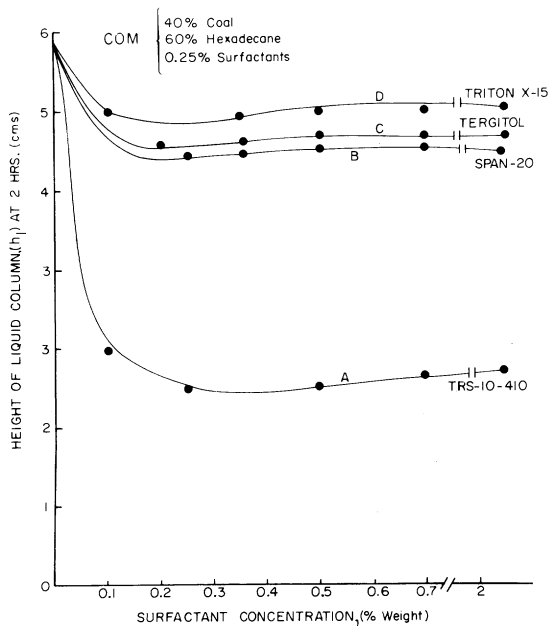Fig. 6. Effect of surfactants on stability of COM.

Fig. 7. Effect of concentration of surfactants on stability of COM.

highest subsidence volume (coal bed). The larger volume of the final settled bed in the case of COM containing surfactants would suggest the network formation. The type of settling observed in these concentrated slurries with or without surfactants could be described as subsidence [12,22].

The effectiveness of these surfactants in reducing viscosity (Fig. 3) and enhancing stability (Fig. 7) above a critical concentration of surfactants suggests that adsorption of the surfactants on coal particles plays a significant role in these phenomena. Also, the fact that surfactants of some degree of effectiveness are found among cationic, anionic and even nonionic classes as shown by Rowell et al. [12] appears to rule out simple electrical (charge neutralization) mechanisms which are essential for stabilization.

Coal—fuel oil No. 6 and coal—fuel oil No. 6—alcohol dispersions

It was found that fuel oil No. 6 and ethanol were not compatible. However, these liquids can be blended together in the presence of an appropriate amount of a third component such as n-alkane or kerosene.

The results of the compatibility study of petroleum liquids and coal derived liquids have been reported recently [13]. The results of compatibility tests of fuel oil No. 6 + ethanol + kerosene are shown in Table 1. It is interesting to note that there are compositions of fuel oil No. 6, ethanol and kerosene which yield compatible mixtures (Table 1). Such compatible alcohol + fuel oil compositions can be referred to as "alcofuels". It should be emphasized that alcofuels exhibit much lower viscosity as compared to fuel oil No. 6 (Table 1).

Figure 8 shows the viscosity of coal dispersions in fuel oil No. 6 and in alcofuel A (Composition shown in Table 2) as a function of coal concentration. It is evident that COMs prepared from alcofuels exhibit much lower

TABLE 1

Compatibility of fuel oil No. 6 with ethanol

Blend composition (wt.%)		Ethanol to kerosene ratio	Incompatible volume		Viscosity (cp)
Fuel oil	Ethanol + kerosene		V_1 (ml)	V_{inc} (%)*	
65	35	10.6	0.2	2.0	
		6.0	0.1	1.0	
		2.5	<0.05	<0.5	30
		1.33	0	0	
70	30	14.0	0.3	3.0	
		0.5	0.1	1.0	
		2.75	<0.05	<0.5	42
		1.0	0	0	
75	25	4.0	0.1	1.0	
		3.0	<0.05	<0.5	50
		1.0	0	0	
100					220

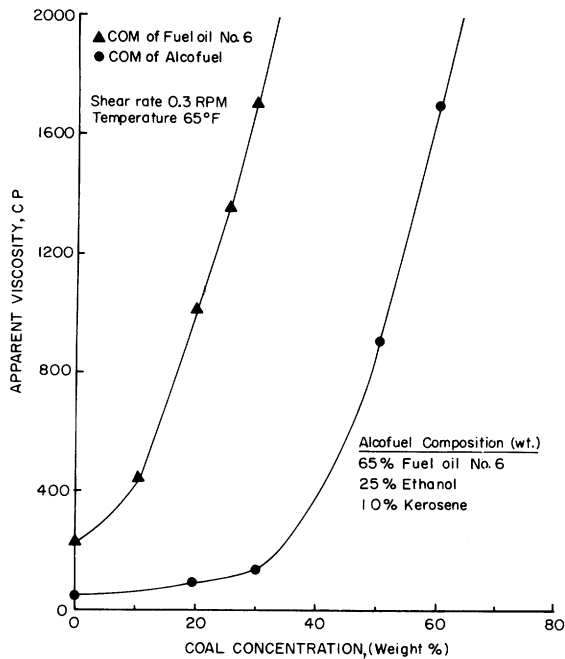*Total volume of blends, V_t = 10 ml in each case.

Fig. 8. Effect of coal concentration on apparent viscosity of COM.

TABLE 2

Heating values of fuel oil No. 6 containing COM and alcofuel COMs

System	Heating values (kJ/g)
Coal	34.30
Fuel oil No. 6	38.60
Coal 25% + fuel oil No. 6, 75%	38.50
Coal 25% + alcofuel A*, 75%	37.86
Coal 25% + alcofuel B**, 75%	37.78

*Alcofuel A contains fuel oil No. 6, 65%, ethanol 25%, kerosene 10% by weight.
**Alcofuel B contains fuel oil No. 6, 75%, ethanol 20%, kerosene 5% by weight.

viscosity than those prepared with fuel oil No. 6. The viscosity of alcofuel containing 35% coal is comparable to that of fuel oil No. 6 ($\approx$ 220 cp). It is evident from Fig. 8 that about 50—60% coal can be added to alcofuel while maintaining pumpable viscosity. The heating values of coal, fuel oil, alcofuels and various coal—oil mixtures are shown in Table 2. The alcofuel COMs exhibit comparable heating values as that of fuel oil No. 6. It appears that the incorporation of alcohol may cause swelling of the basic coal structure resulting in an increased pore volume and surface area. This may bring about

a greater reactivity or combustibility of coal [14]. Thus, we believe that alco-fuels, by themselves as well as with dispersed coal, offer a promising fuel for the immediate future.

Figures 9 and 10 illustrate the present and alcofuel COM technologies. Currently, fuel oil No. 6 is heated to $\approx 150°F$ in order to bring it to an easily flowable condition. Additives are incorporated into the fuel oil and then mixed with the appropriate weight percent of coal in a processing tank, at elevated temperatures. The COM so prepared is stored in a storage tank with continuous agitation to avoid settling of coal particles (at $\approx 150°F$). The COM is heated during pumping and atomization prior to combustion [23,24]. Fig. 9 depicts the flow sheet of current COM technology.
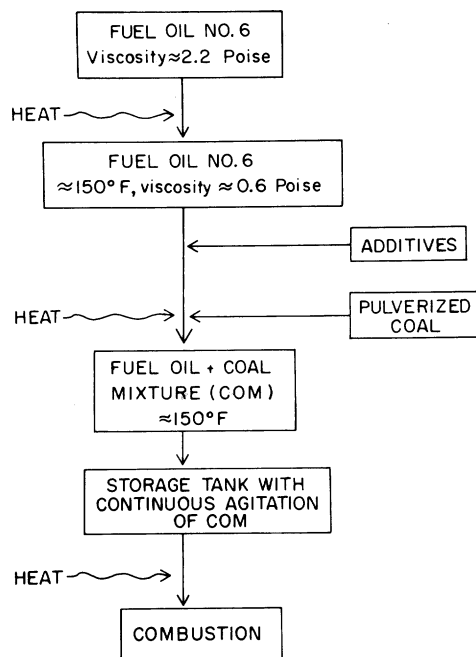
CURRENT STATUS OF COM TECHNOLOGY

Fig. 9. Current status of COM technology.

In alcofuel technology, ethanol and kerosene are mixed in suitable proportions and blended with fuel oil No. 6 at ambient temperature. COM processing is similar to the current method except that heating is not required at any stage due to the lower viscosities of the alcofuel fluids. Fig. 10 illustrates the various steps in alcofuel COM technology. The basic differences between the two technologies are: (1) considerable energy savings by alcofuel COM technology (mixing, storing, pumping at ambient temperature) and (2) use of ethanol as fuel oil extender.

PROPOSED ALCOFUEL COM TECHNOLOGY

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┌──────────────┐   ┌──────────┐   ┌──────────────┐
│ FUEL OIL NO.6│   │ ETHANOL  │   │  KEROSENE    │
│              │   │          │   │     or       │
│              │   │          │   │  n-ALKANE    │
└──────┬───────┘   └────┬─────┘   └──────┬───────┘
       │                │                │
       │           ┌────▼────────────────┘
       │   ┌────────▼──────┐
       └──►│   BLENDED     │◄───
           │   ALCOFUEL    │
           └───────┬───────┘
                   │    ┌──────────────┐
                   │◄───┤  ADDITIVES   │
                   │    └──────────────┘
                   │    ┌──────────────┐
                   │◄───┤  PULVERIZED  │
                   │    │     COAL     │
                   │    └──────────────┘
           ┌───────▼───────┐
           │  COM-ALCOFUEL │
           └───────┬───────┘
                   │
           ┌───────▼───────┐
           │ STORAGE TANK  │
           │ COM-ALCOFUEL  │
           └───────┬───────┘
                   │
           ┌───────▼───────┐
           │  COMBUSTION   │
           └───────────────┘
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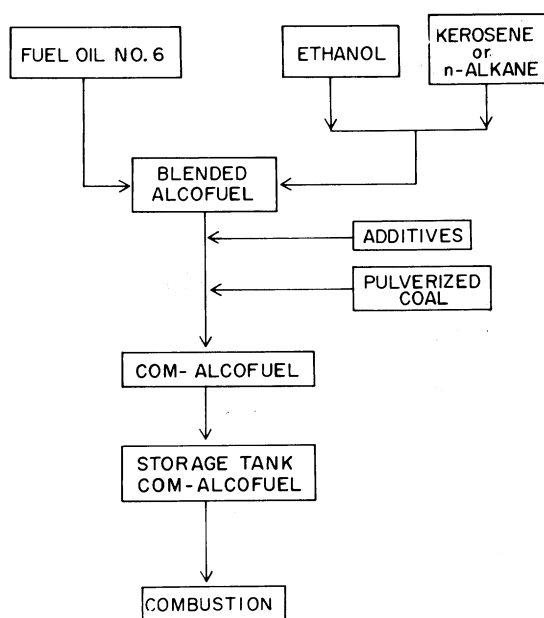
Fig. 10. Proposed alcofuel COM technology.

CONCLUSIONS

It appears from the studies of a model system coal + hexadecane that surfactants such as TRS 10-410, SPAN-20, etc., decrease the apparent viscosity and increase the stability of COM. The apparent viscosity reduction could be due to the adsorbed surfactants which reduce the electroviscous forces between the coal particles. Beyond a specific concentration, further addition of surfactants have hardly any effect on viscosity or stability. The coal + hexadecane dispersions were found to be shear thinning type.

A novel method of blending fuel oil No. 6 and ethanol has been developed using n-alkanes or kerosene as coupling agents. The compatibility tests of fuel oil + ethanol in the presence of kerosene yield a range of compositions which are compatible. The compatible blends (alcofuels) have considerably lower viscosities than fuel oil No. 6 and the COMs containing alcofuels and 60% coal have comparable viscosities as that of COM containing fuel oil No. 6 and 20% coal. The coal + oil mixtures prepared with fuel oil No. 6 and with alcofuels have similar heating values. The advantages of alcofuels as compared to those of fuel oil No. 6 are emphasized in relation to COM technology.

ACKNOWLEDGEMENT

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