Fuel from Microalgae Lipid Products

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ABSTRACT

The large-scale production of microalgae is a promising method of producing a renewable feedstock for a wide variety of fuel products currently refined from crude petroleum. These microalgae-derived products include lipid extraction products (triglycerides, fatty acids, and hydrocarbons) and catalytic conversion products (paraffins and olefins).

Microalgal biomass productivity and lipid composition of current experimental systems are estimated at 66.0 metric tons per hectare year and 30% lipid content. Similar yields in a large-scale facility indicate that production costs are approximately six times higher than the average domestic price for crude, well-head petroleum. Based on achievable targets for productivity and production costs, the potential for microalgae as a fuel feedstock is presented in context with selected process refining routes and is compared with conventional and alternative feedstocks (e.g., oilseeds) with which microalgae must compete.

INTRODUCTION

Lipid oils derived from microalgae have been the focus of considerable interest because these oils contain fatty acid and triglyceride compounds that can be esterified into alcohol esters using conventional technology. The resulting so-called ester fuels have proven suitable for blending with diesel stocks up to 30% without affecting engine performance [1], and some investigators have successfully used them as neat fuels [2,3].

Microalgae are unique, photosynthetically driven chemical factories. More than 40 years of research in microalgae has demonstrated that these organisms produce a diverse array of chemicals and hydrocarbon fuels. These products offer promise as potential substitutes for products currently derived from petroleum and natural gas. Figure 1 is a schematic overview of microalgal fuel processing routes and products. Microalgal-derived products include extraction products such as hydrocarbons, fatty acids, glycerol, protein, and polysaccharides [4,5,6]; bioconversion products such as alcohols, organic acids, and methane [5,7]; and catalytic conversion products such as paraffins, olefins, and aromatics [7,8,9]. Microalgae could be attractive feedstocks for conversion to any of these products if their productivity and versatile chemical composition can be exploited.

This paper discusses microalgae as a feedstock for one process identified in Figure 1—the production of oxygenated fuels suitable for supplementing existing supplies of refined hydrocarbon fuels from conventional sources. This paper also presents a review of the commercial and developmental uses
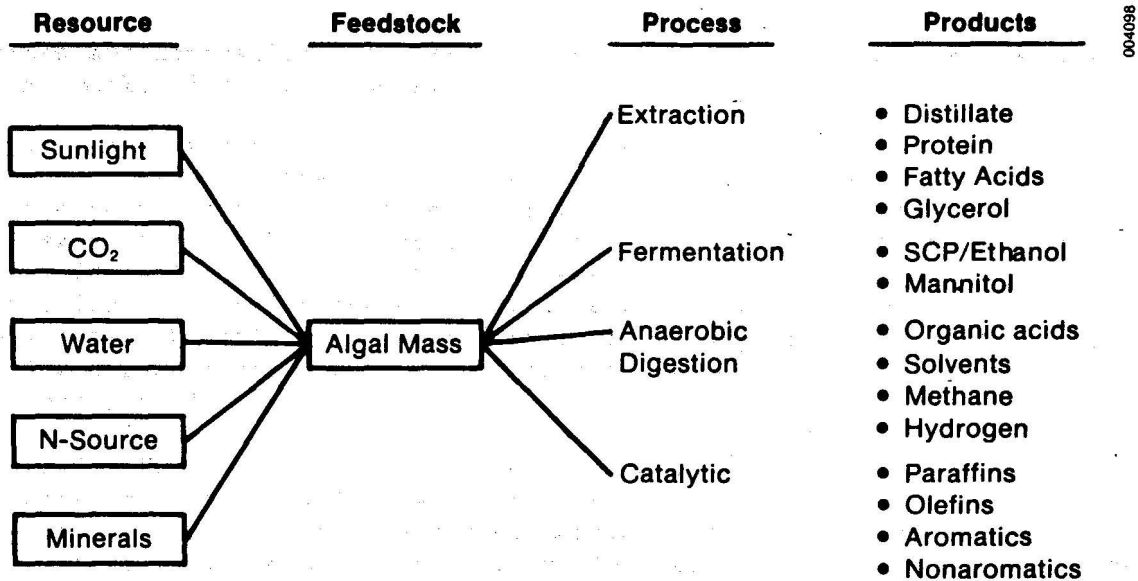
Resource	Feedstock	Process	Products

Figure 1. Schematic of Microalgae Processing Options for Fuel Products

of transesterification for conversion of algal biomass into high-quality liquid fuels. The products of transesterification are compared with conventional (diesel) and nonconventional (vegetable oil) fuels and assessed for their suitability as fuel products.

TRANSESTERIFICATION

Transesterification is the conversion of glycerol esters (triglycerides) to monoesters of lower alcohols, typically methanol or ethanol, as shown in Figure 2. This process is commonly used in the detergent industry for the conversion of triglycerides into fatty acids, esters, and alcohols. While either acids or bases have been used as catalysts, bases such as sodium hydroxide (caustic soda) and sodium methoxide have produced slightly faster conversions at higher conversion rates than the acids. Of the wide range of
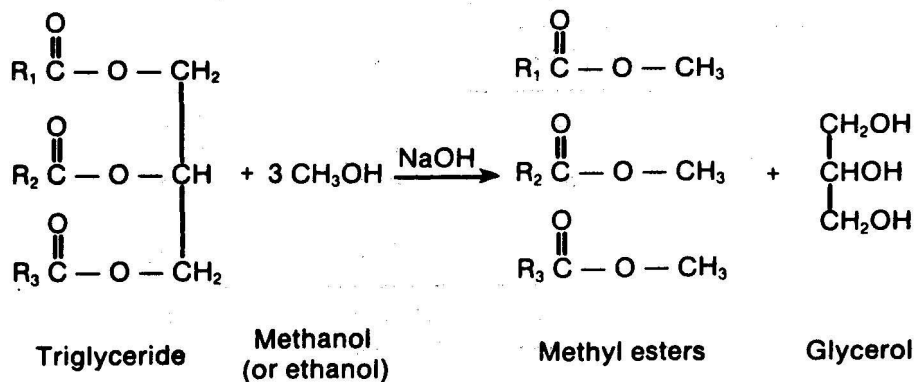
$R_1, R_2, R_3 = 10$ to 30 carbons

Figure 2. The Transesterification Reaction

temperatures investigated, 45°-50°C seems to be the range at which the rates begin to increase noticeably [10,11,12]. Reactions at 60°C have proved quite successful (95%-98% yields have been reported), while yields at 80°-100°C have been in the 80%-90% range..

Figure 3 presents the adaptation of a typical transesterification process to microalgae. It is assumed that some preliminary treatment of the algae such as harvesting and drying would be necessary, but the exact conditions cannot yet be specified. Extraction with a suitable solvent would produce a reactor feed similar to the vegetable-seed oils that are already used for the process. A mixture of chloroform and methanol, which is typically used in total lipid determinations, might be used [13]. Both these solvents boil at 60°-65°C and form a minimum-boiling azeotrope at 53°C at atmospheric pressure, so solvent stripping would be readily accomplished. Transesterification is typically carried out in jacketed, agitated reactors. Molar alcohol-to-feed ratios of 5:1 and up, considerably in excess of the 3:1 stoichiometric ratio, are required for high product yields. By-product glycerol, which is considerably higher in density, forms a heavier liquid phase and is easily separated. The product stream is then neutralized, the unreacted alcohol is recovered for reuse, and the ester product is suitably purified.

THE POTENTIAL FOR THE USE OF ESTER FUELS

Research interest in the transesterification process is due to the market demand for diesel fuel and its relatively high cost as a refined fuel product. Domestic demand for distillate (e.g., No. 2 diesel) in 1981 was 2.84×10^6 bbl/d, representing 6 quads of energy consumed annually. The transportation sector's share of the demand for this distillate is 41.0% of total consumption (2.47 quads); the balance is required by stationary
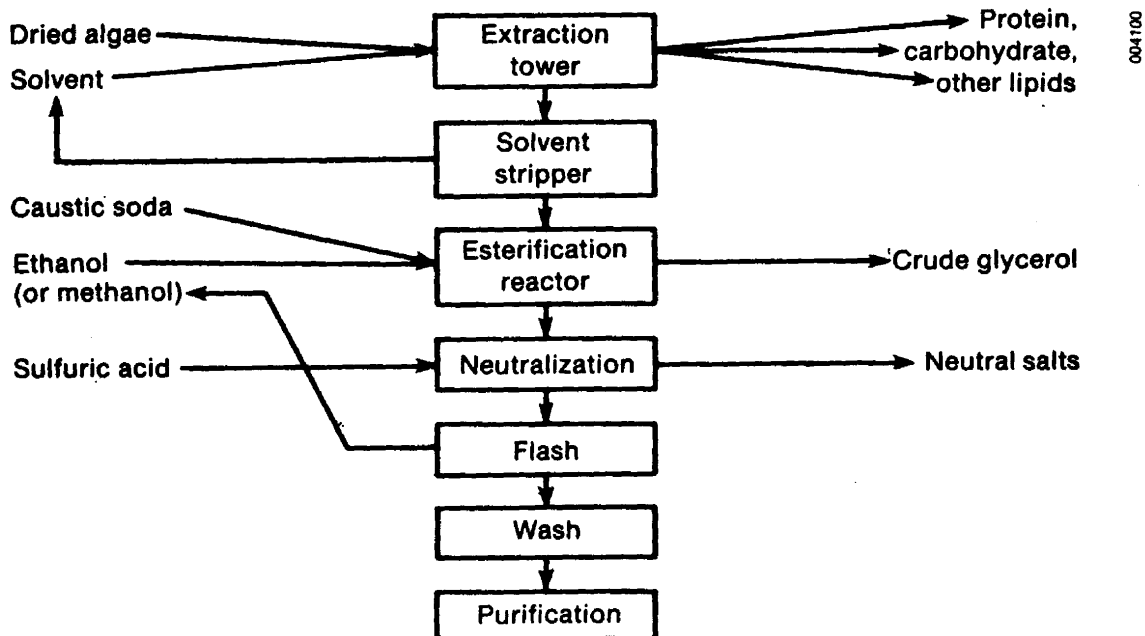

Figure 3. Conceptual Transesterification Process Using Microalgae as a Feedstock

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engines and combustion furnaces. These estimates of demand represent a high volume market for ester fuels.

However, the relatively low cost and convenience of No. 2 diesel fuel has prevented any significant development of alternative fuels such as ester fuels except during constrained supply situations similar to the petroleum shortages of World War II and the early 1970s. In the latter situation, escalating prices for conventional fuels generated sufficient interest in developing alternative fuels that a recent worldwide survey of oxygenated fuels research activities identified over 100 organizations that are conducting engine testing experiments to further develop the utilization of oxygenated fuels [14].

Prices for No. 2 diesel fuel to the year 2000 have been estimated based on projected prices for well-head crude petroleum and several simplifying assumptions regarding refinery cost margins for petroleum products such as No. 2 diesel. These price projections for petroleum establish a relative cost of ester fuels ($/gal) that would enable them to be cost-competitive with diesel fuel in the future. The simplifying assumptions are necessary because actual refinery operations and pricing strategies are complex and dynamic. Table 1 provides EIA price projections for crude petroleum to the year 2000. By 2000, the high price projection is $80 (in 1983 $) per barrel of crude. It is assumed that additional refinery acquisition and transportation costs for No. 2 diesel oil may increase product prices by 35% [15]. If this cost margin remains constant to the year 2000, the projected price for No. 2 diesel would be $2.75/gal (in 1983 $). The high price and high market demand for diesel projected for the year 2000 provide opportunities for renewable fuels to be developed as potential diesel substitutes. However, it is important to compare the properties of the desired diesel fuel with the various substitute fuel products.

Table 1. World Oil Price Projections, 1983 to 2000[a]
(1983 dollars per barrel)

Year	Oil Price Scenario		
	Low	Middle	High
1983	29.0	31.0	33.0
1985	32.0	26.0	36.0
1990	29.0	39.0	50.0
1995	39.0	58.0	76.0
2000	44.0	62.0	86.0

[a]All prices reflect the average landed price of crude oil in the United States.

Source: U.S. Department of Energy, Energy Information Administration, April 1983, Annual Energy Outlook with Projections to 1990, DOE/EIA-0383 (83).

Figure 4 compares the gross heating values of a given volume of No. 2 diesel fuel, crude sunflower oil, and the methyl ester of sunflower oil. The vegetable oil has about 95% of the fuel value of diesel, while the methyl ester has about 90% [16]. Either the vegetable oil or methyl ester could be an acceptable diesel substitute under a variety of conditions on its fuel value alone.

Another critical diesel fuel property is the cetane number (CN), a numerical indication of its ignition quality. The cetane number is related to such engine performance factors as ease of starting, warmup, combusion roughness, acceleration, deposits, and exhaust smoke. A fuel of higher cetane number has a shorter ignition delay period, is easier to start, and has little or no combusion roughness [17,19].

Figure 5 shows the range of cetane numbers found in diesel, vegetable oils, and vegetable oil methylesters [16,18]. The cetane numbers shown for vegetable oil are thus barely acceptable. Transesterification, however, causes a marked improvement, increasing values to higher than diesel itself. This could be especially important in light of the gradual but steady decline in the overall cetane number of the diesel stock that began in the late 1960s and has accelerated since 1976, primarily due to the refining industry's increasing use of heavier crudes that require more severe cracking and yield distillates with appreciably lower cetane numbers.

The next physical property to be considered is viscosity; Figure 6 shows viscosity for No. 2 diesel, vegetable oils, and vegetable oil esters. The viscosity of vegetable oils at 40°C is typically higher than either diesel or ester fuel by a factor of ten, and increases much more sharply with decreasing temperature. Viscosity of vegetable oil at 0°C is over 20 times higher than its esters or diesel fuel [16,18]. The high viscosity can
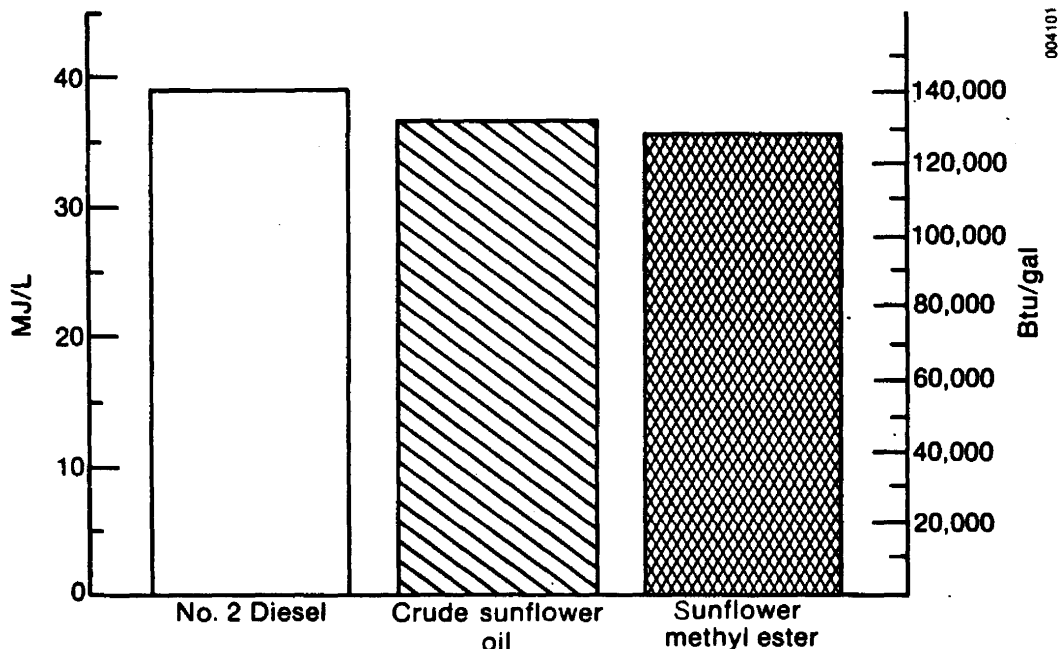
Figure 4. Diesel Fuel and Diesel Substitutes: Gross Heat Value (volumetric basis)
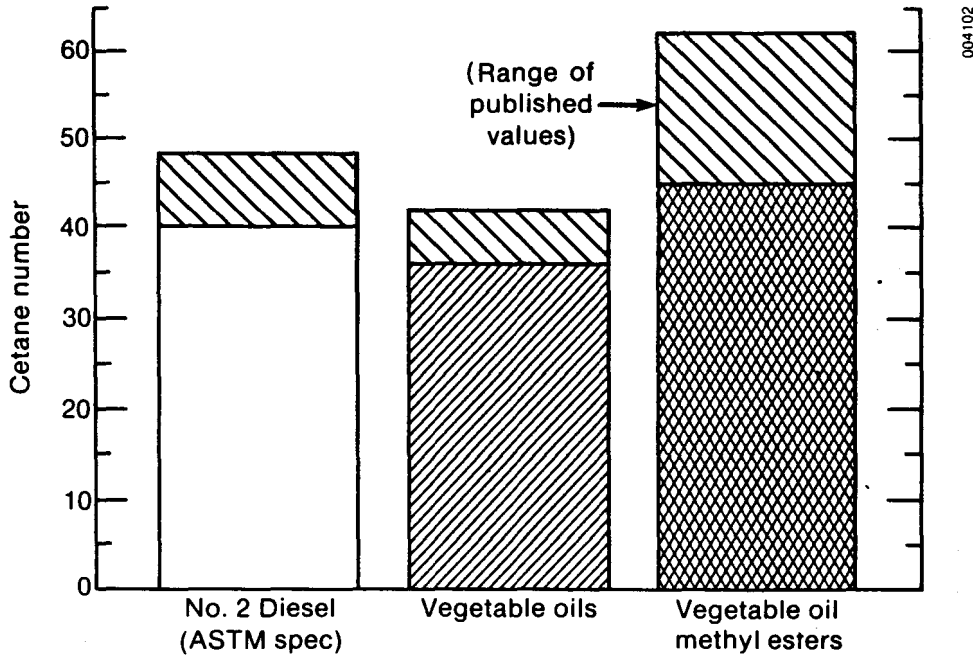
Figure 5. Diesel Fuel and Diesel Substitutes: Cetane Number
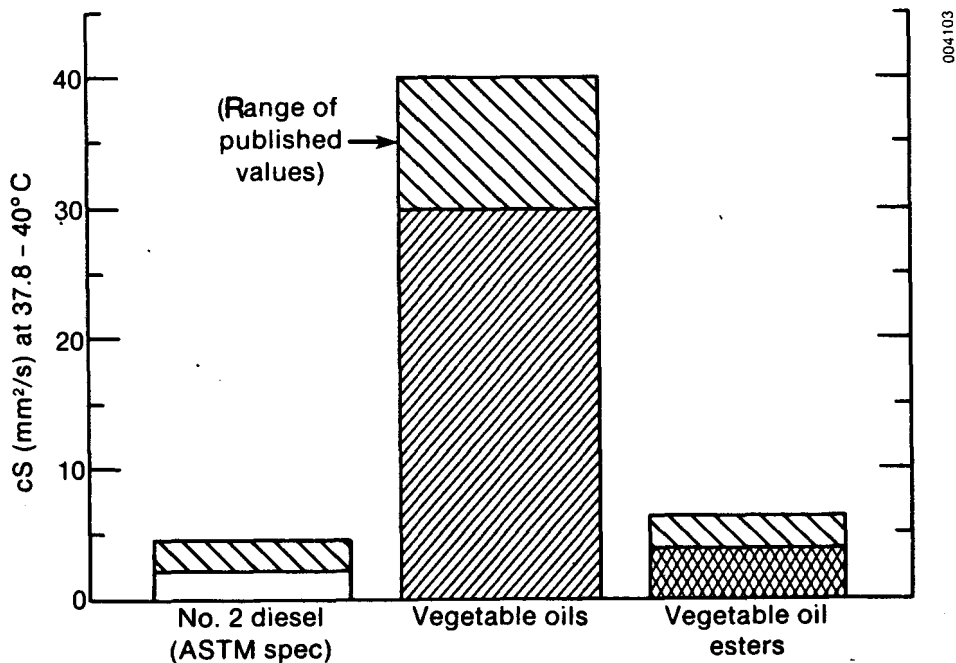
Figure 6. Diesel Fuel and Diesel Substitutes: Viscosity

increase injector fouling, cause carbon deposits on cylinder walls and exhaust ports, and cause fuel system problems such as filter plugging. The ester fuel's viscosity is within the industry-specified range for diesel fuel.

Another important property is the cloud point. This is the temperature at which, upon cooling, the appearance of haziness or cloudiness is first

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observed. The cloud point is generally considered the lowest temperature to which a fuel should be subjected. Figure 7 shows that neither the oils nor the esters have cloud points as low as diesel fuel, but oils have a slight advantage over the esters in this regard. With unheated fuel lines, the ester's 0°-10°C cloud point makes it unsuitable for winter use, but above 10°C (50°F), in a heated system, or in indoor boilers or generators it would perform acceptably.

The preceding discussion highlights several of the technical considerations important in developing suitable diesel fuel substitutes from biomass resources. It is important that these renewable fuels closely match the characteristics of diesel fuel in order to minimize adverse diesel engine performance or the need for extensive engine modification.

Manipulation of the fatty acid composition of microalgae-derived feedstocks for transesterification may be accomplished by modifying environmental conditions within a culture system or by changing the species cultured. In Figure 8 the fatty acid composition for two species of microalgae is shown relative to the fatty acid levels of sunflower oil [13,20]. The most significant difference between the microalgae fatty acids and those of sunflower oil is the large amount of linoleic acid (18:2) in sunflower oil. The microalgal species Ankistrodesmus and Isochrysis show a higher degree of unsaturation (16:2+, 18:3, 20+:1+) in the longer chained fatty acids, higher concentration in shorter chained saturated fatty acids (14:0, 16:0), and overall a much greater variety of acids. The two lines of data for Isochrysis in Figure 8 represent the algal growth (a) with sufficient nitrogen, and (b) under nitrogen-deficient conditions. The difference in the fatty acid profiles for Isochrysis is striking because of environmentally created shifts in the fatty acid composition. Under nitrogen-deficient conditions, the algae actually produce less palmitic acid than sunflower oil.
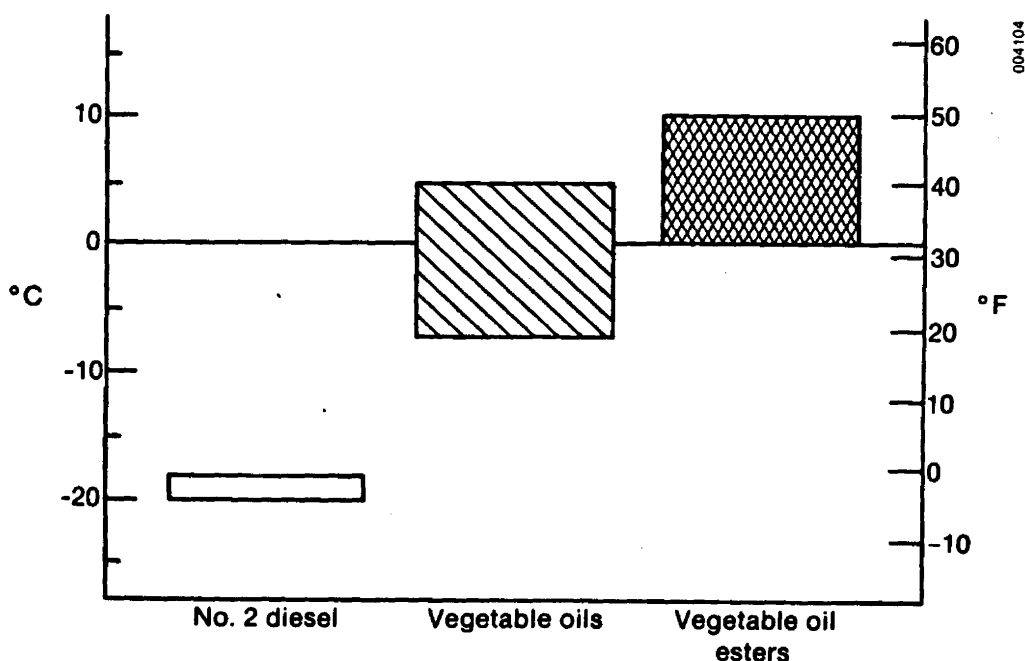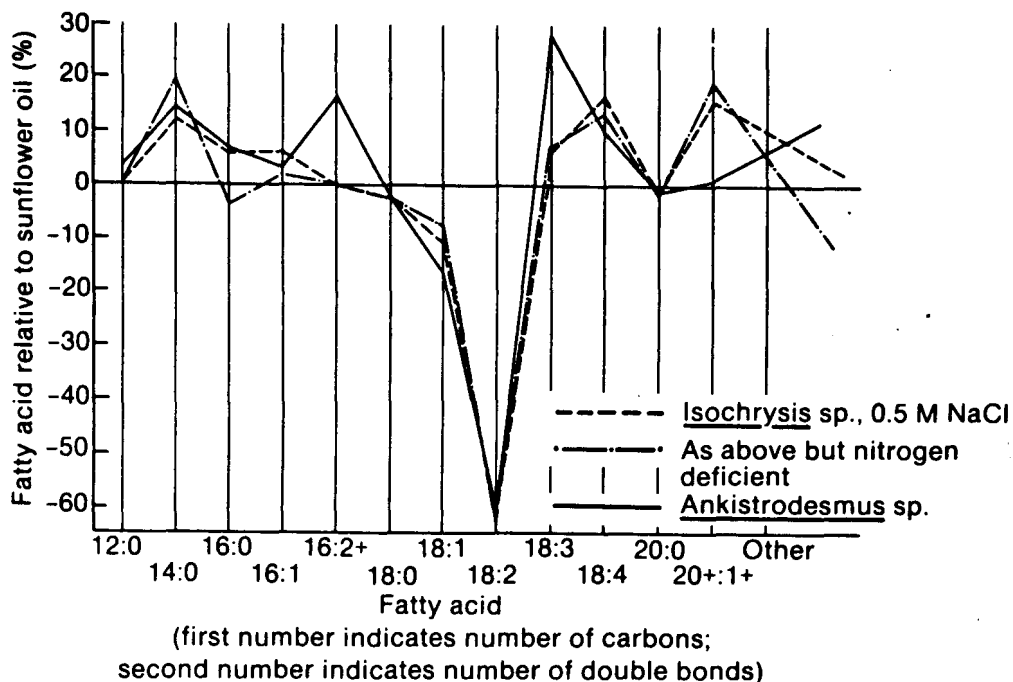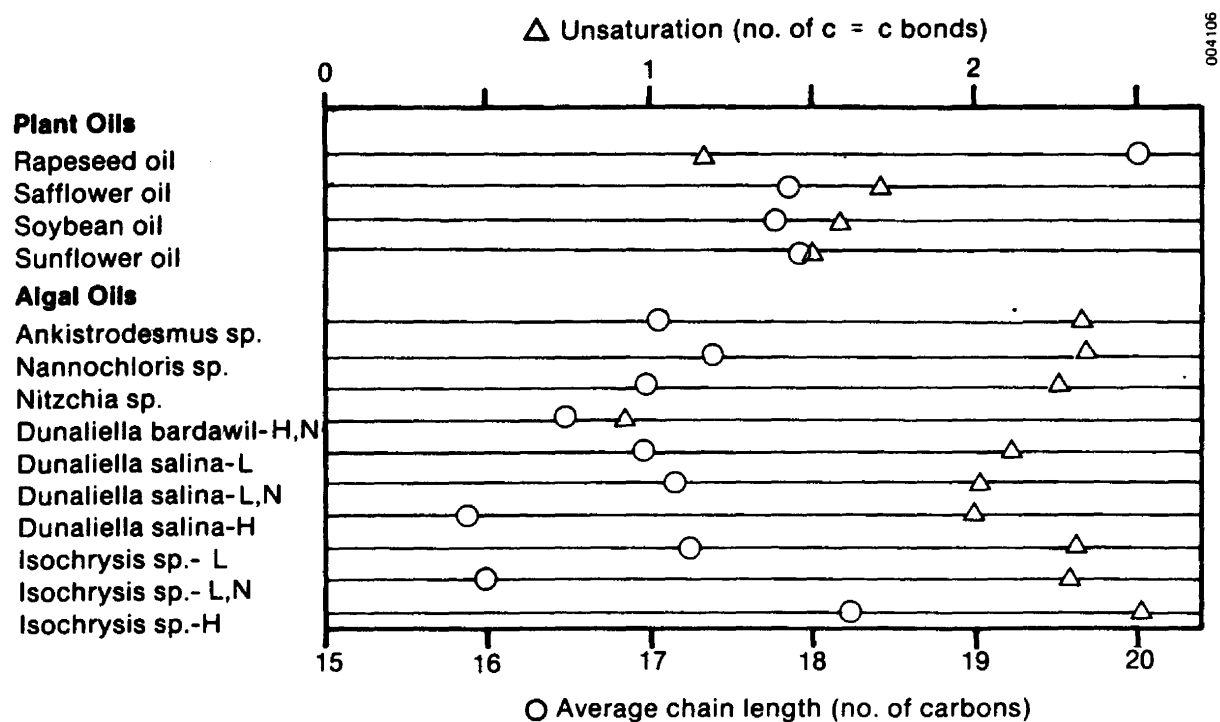

Figure 7. Diesel Fuel and Diesel Substitutes: Cloud Point

Figure 8. Algal Lipid Profiles (relative to sunflower oil)

A similar comparison of fatty acid profiles of vegetable oils to microalgae oils is shown in Figure 9. The different vegetable oils are closely grouped in terms of degree of unsaturation and average chain length of all fatty acids present in the oils. Microalgae oils show a greater range of variability between species and within species grown under different environmental conditions. This figure, like Figure 8, supports the general conclusion that microalgae fatty acids tend to be more unsaturated and have shorter carbon chains. Yet, environmental modification of the culture conditions shows that wide swings are possible in both degree of unsaturation (Dunaliella) and in carbon chain length (Isochrysis).

These fatty acid compositional changes, available in microalgae, are important features in developing suitable feedstock qualities for esterification. The desired properties of the final ester fuel product (e.g., viscosity, cloud point, oxidation, etc.) are directly related to the chemical properties (e.g., unsaturation and carbon chain length) of the feedstock. The demonstrated flexibility to significantly alter the fatty acid profiles of microalgae oils provides opportunities to mitigate adverse fuel product characteristics by growing the appropriate microalgae species under specific growth conditions to achieve maximum values of desired fatty acids. Specifically, reductions in carbon chain length would reduce the melting point, which in turn would lower the cloud point and the viscosity. Minimizing double bonds (unsaturation) lowers the susceptibility to oxidative polymerization, thus lengthening storage life and minimizing both wax formation and lubricant contamination. Any of these improvements that can be made during algal production serve to reduce associated refining costs necessary to upgrade the quality of the final fuel product (e.g., hydrogenation to increase chemical stability to oxidation).

Δ Unsaturation (no. of c = c bonds)

004106

Plant Oils
Rapeseed oil
Safflower oil
Soybean oil
Sunflower oil

Algal Oils
Ankistrodesmus sp.
Nannochloris sp.
Nitzchia sp.
Dunaliella bardawil-H,N
Dunaliella salina-L
Dunaliella salina-L,N
Dunaliella salina-H
Isochrysis sp.- L
Isochrysis sp.- L,N
Isochrysis sp.-H

O Average chain length (no. of carbons)

Key
L — Grown under low salinity (0.5 M NaCl)
H — Grown under high salinity (1-2 M NaCl)
N — Nitrogen deficient

Figure 9. Fatty Acid Composition of Various Algae and Plant Oils

TRANSESTERIFICATION FEEDSTOCKS AND YIELDS

The amount of resource available for transesterification depends on two factors—biomass yield and lipid content. Biomass yield provides a measure of photosynthetic efficiency in converting sunlight into organic matter. Lipid content, as a fraction of total biomass, represents the amount of hydrocarbons and triglycerides formed as storage products resulting from photosynthetic metabolism. A comparison of reported biomass and triglyceride yields for sunflower and microalgae is presented in Table 2.

Duke and Bagby [21] reported that sunflower biomass yield varies from 4 to 9 t/ha* with seed yields ranging from 300 to 3000 kg/ha. The highest average commercial sunflower seed yield in the United States is 1267 kg/ha; based on a 40%-48% oil content, this yield provides 506-608 kg/ha of oil. Experimental plantings of sunflowers have shown oil yields as high as 1543 kg/ha based on a crop yield of 3181 kg/ha and a 48.5% oil content. These experimental plantings yield 10 barrels of sunflower oil per hectare, which is more than double the oil yield based on the 1979-80 average yield of sunflowers in the United States (1326 kg/ha with a 40% oil content) [22].

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*Metric ton (1000 kg) is abbreviated "t" throughout this paper.

Table 2. Biomass and Triglyceride Product Yield Comparison Between Microalgae and Sunflower

Source	Biomass Yield (kg/ha yr)	Oil Content (%)	Oil Yield (bbl/ha yr)
Sunflower (commercial)	1,326	40.0	3.5
Sunflower (research)	3,181	48.5	. 10.5
Microalgae (commercial)	66,000	15.7[a]	62.0
Microalgae (research)	124,000	38.4[a]	325.0

Sources: Sunflower yield data from Ref. 21 and Ref. 22; microalgae yield data from Ref. 23.

[a]Oil content calculated as 0.644 x total lipids.

Microalgae yields are reported as net productivities in dry, ash-free grams per square meter per day. Based on the current results of field experiments, net productivity of microalgae is 23 g m^{-2} d^{-1}. After harvesting, net yield of microalgae is 66.0 t/ha yr, assuming 5% harvesting losses and a 300 d/yr operation.

Depending on the species and growth conditions, chemical analysis of microalgae has shown that 10%-70% of the dry ash-free weight is comprised of lipids. The outdoor field experiments with the microalgae Ankistrodesmus have achieved a total lipid content of 24.5% of the total organic weight.

The data in Table 2 are distinguished by two estimates that represent commercial yields and higher yields indicative of reported research results for sunflower and microalgae. The research yields provide indications of the potential improvements that may be achieved with careful cultivation practices and strain improvements. Research has produced significantly greater yields—300% improvement for sunflower and 524% for microalgae—than those available commercially. Table 2 shows that microalgae offer a significant yield advantage over sunflower as a potential feedstock for transesterification. The percentage of the total lipid component available for esterification was estimated to be 64.4% and used to calculate the oil yields from microalgae shown in Table 2.

MICROALGAE PRODUCTION ECONOMICS

Recent economic analyses of microalgae cultivation indicate that a 405-ha facility producing 66.0 t/ha yr (23.0 g m^{-2} d^{-1}) has a levelized annual cost of $11.1 million (1983 $) [23]. Unit cost under this production scenario is $415/t of algae. The cost breakdown for this facility is presented in Table 3.

Installed costs for the growth ponds, pipes, pumps, buildings, offsites, and harvesting system are estimated to be approximately $55,330/ha. Additional capital cost charges for engineering, contingencies, and land result in total fixed costs for the microalgae production facility of $75,560/ha. On

an annualized cost basis, these fixed costs ($5,150/ha) represent 18.8% of the total costs for microalgae production as indicated in Table 3.

Variable costs—costs associated with annual operation of the microalgae facility—represent 81.2% of the total annualized costs and are directly proportional to production output. The major variable cost is nutrients (e.g., carbon and nitrogen) required to sustain the biomass production (54.5% of the total variable costs). Utilities required to run the pumping subsystems (e.g., recirculation) constitute the second largest. operating cost category. The total annualized variable costs for the system are $8.9 million or $336.5 per metric ton of biomass.

Financial assumptions include a 50% debt ratio, 35% common stock, and 15% preferred stock at real rates of return of 3.7%, 5.0%, and 4%, respectively. The facility's operating lifetime is 30 years with a capital depreciation schedule of 15 years. Real escalation rates of 1.0%/yr for capital costs and 1.5%/yr for operating costs are assumed. The real weighted average after-tax cost of capital of 3.4% represents the facility's internal rate of return based on a revenue required cost methodology.

The "commercial" microalgae unit cost estimates (presented in Table 3) are twice the commercial market value of sunflower seed. This cost disadvantage would make microalgae a less likely transesterification feedstock than sunflower seeds. Other noneconomic factors would have to be considered to make microalgae a viable feestock for transesterification. These noneconomic factors would include microalgae's capability to utilize marginal resources (e.g., arid lands and brackish water) and their high productivity (see Table 2).

Table 3. Annualized Cost Breakdown for "Commercial" and "Improved" 405-ha Microalgae Facilities (1983 $)

	Dry Biomass ($/t)	
	Commercial[a]	Improved[b]
Annualized Capital Investment	78.0	48.2
Annual Operating Cost		
Utilities	56.0	35.5
Nutrients	226.0	108.6
Operation and maintenance	21.0	13.1
Labor and overhead	34.0	21.6
Total production cost	415.0	227.0

[a]Based on a 23 g m^{-2} d^{-1} productivity (66 t ha^{-1} yr^{-1}).

[b]Based on 43 g m^{-2} d^{-1} productivity (124 t ha^{-1} yr^{-1}).

The two reasons that microalgae should be considered as a feedstock for transesterification are the potential for significant yield improvements through continued research and the potential ability to manipulate chemical composition of microalgae to maximize certain fatty acids. As Table 2 indicates, microalgae could achieve biomass yields of 124 t ha^{-1} yr^{-1} (43.4 g m^{-2} d^{-1}) with a fatty acid yield over 320 bbl ha^{-1} yr^{-1}. Levelized total annual cost for a 405-ha microalgae facility at these improved production levels is \$11.40 million (1983) or \$227.0 t^{-1} of biomass. This analysis assumes an improved facility over the commercial facility in algal productivity (23 to 43 g m^{-2} d^{-1}), lipid content (30% to 60%), 90% recycling of noncarbon nutrients, and lower carbon dioxide prices (\$82.5 to \$38.5 t^{-1}).

Comparing the two sets of microalgae production costs in Table 3 shows that the ability to achieve the improvements in specific biological and engineering performance levels results in cutting unit product costs for microalgae in half. Because microalgae are potential feedstocks for esterification, this cost reduction is significant and offers competitive potential when compared with sunflower seed oil.

Table 4 illustrates the unit cost (\$/gal) comparison between sunflower oil and microalgae oil as a fuel or as a transesterification feedstock. The biomass costs for the two microalgae facilities in Table 3 are converted to dollars per gallon. The sunflower unit cost estimates are adapted from Lipinsky's study of producing vegetable oils from various seed crops as an emergency diesel fuel [22]. The estimates from Lipinsky's study are for a commercial venture subject to the financing assumptions previously described in this paper.

By-product credits from sunflower oil seed expelling and microalgae processing were excluded from the estimates presented in Table 4. Each facility would have a large product stream of protein feed available for selling after processing; however, determining these by-products credits is beyond the scope of this paper and clouds the issues of specifying production costs for transesterification feedstock oils from these renewable resources.

Of the estimates presented in Table 4, the unit costs of the lipid oils from sunflower and the baseline microalgae facilities exceed the projected prices of diesel fuel for the year 2000. Thus, as a neat fuel these products are not cost competitive. Conversion costs to produce methyl esters have been estimated at \$1.41/gal [10] excluding feedstock costs. Adding to esterification costs the feedstock costs associated with sunflower and microalgae, a methyl ester fuel from these renewable feedstock would cost a minimum of \$4.50/gal excluding by-product credits. At the higher microalgae yields the ester fuel would cost \$3.50, which is also above the projected future price of diesel fuel. However, the higher cetane numbers of methyl ester fuels offer an opportunity to improve the declining cetane pool value in conventional diesel supplies by using methyl ester fuel as a cetane enhancer, which is similar to use of ethanol as an octane enhancer for gasoline.

CONCLUSIONS

The high productivity and diverse chemical composition make microalgae a unique and promising feedstock for conversion into a variety of liquid and

Table 4. Comparison of Production Costs for a Sunflower and Microalgae as Feedstocks to Transesterification

	Commercial Sunflower Oil Processes	Microalgae Oil Processes	
		Commercial	Improved Research
Capacity (gal/yr)	0.589×10^6 [a]	3.65×10^6 [b]	13.71×10^6 [c]
Cost Parameters ($/gal)			
Capital cost	0.04	1.41	0.436
Operating cost			
Utilities	0.15	1.01	0.32
Maintenance	0.07	0.38	0.11
Labor and material	0.15	0.61	0.19
Subtotal	0.36	2.00	0.62
Feedstock	2.76[d]	2.09[e]	0.98[f]
Production cost ($/gal, volume basis)	3.16	7.50	2.03

[a]Adapted from Ref. 22.

[b]Based on 23 g m^{-2} d^{-1}.

[c]Based on 43 g m^{-2} d^{-1}.

[d]Sunflower seed costs at $220/metric ton.

[e]Nutrient expenses required to produce microalgae.

gaseous fuel products. Fuel processes have been identified that utilize extraction, bioconversion, or catalytic conversion steps with microalgae as a feedstock. The inherent diversity in chemical composition of microalgae, determined by species strain and environmental culture conditions, permits commercial operators to select and optimize feedstock characteristics to maximize conversion efficiency and fuel product slates [24].

One fuel process, transesterification, converts microalgal lipids into alcohol esters suitable for blending in amounts up to 30% with No. 2 distillate with no adverse effect on compression-ignition engines. Concerns regarding engine performance that result from higher viscosities, chemical instability, and limited volatility of neat vegetable oil or ester fuels are somewhat abated when these fuels are blended with No. 2 diesel fuel.

The potential of microalgae as a feedstock, similar to sunflower seed, for transesterification depends on continued biological improvements in algal productivity and lipid content. At current biological performance levels, microalgae oils are prohibitively expensive ($7.50/gal) when compared with

sunflower seed oil ($3.16/gal) either as a neat fuel or as a feedstock for transesterification. However, continued research in achieving higher sustainable microalgae yields and lipid content is anticipated to improve the economic feasibility of microalgae as an ester fuel feedstock.

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