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Biodegradability testing of synthetic ester lubricants—effects of additives and usage

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Abstract

The optimised biodegradability test system "O_2/CO_2 Headspace Test with GC-TCD" is used for the assessment of synthetic ester lubricants. The effects of both additives and usage on biodegradability are examined and discussed. Ester based cutting fluids and hydraulic fluids with and without additives are used under defined conditions at machine tools and hydraulic and plain bearing test benches. The lubricants are characterised additionally with respect to kinematic viscosity, acidity and elemental composition. Furthermore, a formulated mineral oil is characterised before and after usage at an hydraulic test bench.

The results clearly show that the mineral oil is far less biodegradable than the ester oils and that their biodegradability is not affected by usage. Biodegradability of the ester oils is mainly depending on the characteristics of the base fluids and not affected by the additives. Antioxidants are influencing stability respectively biodegradability indirectly, since they prevent oxopolymerisation effects. Other effects of usage on biodegradation are not detected. In this context, the antioxidants ensure ready biodegradability and have a positive effect on the environmental fate of synthetic ester lubricants.

Keywords: O_2/CO_2 Headspace Test with GC-TCD; Ultimate aerobic biodegradation; Machine tools; Hydraulics; Plain bearing test bench

1. Introduction

Thirteen percent (EC countries) and 32% (USA) of all used lubricants are emitted into the environment more or less unchanged in properties and appearance (Bartz, 1998). Most of these lubricants are used in loss lubrication frictional contacts and in circulation systems, which are disposed and not collected. Additionally, lubricants are emitted from leaks, and significant amounts remaining in filters and containers or empties have to be taken into account (Bartz, 1998). Hydraulic systems are operating under high pressure. Therefore, large amounts of hydraulic fluids can be released into the environment from leaks or failures of hoses, seals or cylinders (Battersby, 2000).

For these reasons, the distribution, biodegradability and toxicity of lubricants are important factors with respect to sustainable environmental development. On the other hand, the lubricants should be stable during usage under different operating conditions. Generally, lubricants change their chemical and physical fluid properties due to usage. Since the lubricant is a factor which considerably influences the economic as well as the ecological balance of a machine, the analysis of the

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aging behavior is extremely important. A characterisation of the behavior of the properties of lubricants as a result of utilisation is thus the basis for the development of new lubricants.

Biodegradability is the most important aspect with regard to the environmental fate of a substance. It must be tested e.g. for the EU dangerous preparation directive, the eco-labels and the German water hazard class classification (Willing, 1999). For this purpose standardised methods are being used, which ensure comparable and reproducible results in different laboratories. There are even a lot of standardised biodegradability test systems like OECD methods (OECD, 1992) and ISO standard methods (ISO/TR 15462, 1997). A review of these test systems is given by Pagga (1997).

Lubricants are consisting of base fluids and performance enhancing additives. Mineral oils, rapeseed oils and synthetic or native esters and other organic compounds are used as base fluids. Esters are readily biodegradable in contrast to mineral oils. The concentrations of additives usually are below 10% w/w. For the eco-label "blue angel" it is tolerated, that the additives are only potentially degradable, if their concentration is below 7% w/w (RAL-UZ 79, 1996).

It is not generally recommended to test the biodegradability of used lubricants, although they alter significantly during usage. This may be caused by intake of metals, water or air as well as high pressure or temperature. The changes are strongly depending on the operating conditions. Examinations with the CEC-test have shown a decrease of biodegradability of synthetic ester lubricants during usage, although it still was superior to that of mineral oils (Schuelert and Bernhard, 1995). Remmele and Widmann (1998) have tested hydraulic fluids based on rapeseed oil in agricultural machinery with CEC-test and Zahn–Wellens-test. Only in a few cases biodegradability was not satisfying. In most cases it was extensive. But both the CEC-test and the Zahn–Wellens-test are not suitable for biodegradability testing of ester based lubricants.

In this work the newly established O_2/CO_2 Headspace Test with GC-TCD is applied for the characterisation of biodegradability of environmentally acceptable lubricants based on synthetic and oleochemical esters. Effects of additives and usage are demonstrated.

2. Materials and methods

2.1. Chemicals and lubricants

Analytical grade chemicals are used being obtained from Merck KGaA (Darmstadt, Germany) and from Sigma-Aldrich Chemie GmbH (Steinheim, Germany). The tested lubricants for hydraulic and cutting systems are purchasable and established. The base substances of the cutting fluid are di(2-ethylhexyl)adipate and isopropylpalmitate. The base substances of the synthetic hydraulic fluid are di(2-ethylhexyl)adipate and trimethylolpropane–trioleate with native fatty acid composition. The additive composition (antioxidants and extreme pressure/anti-wear substances) is unknown.

2.2. Test benches

The degree of the changes due to usage of lubricants corresponds to the loads applied to the fluids. The investigation of aging thus has to take into account the various specific structures of the tribosystems. Since the investigations are carried out within the framework of the collaborative research centre SFB 442, tribosystems as being used in a machine tool are regarded. So, fluids used in a hydraulic system as well as in plain bearings (Table 1) are analysed with respect to the change of viscosity, the total acid number (TAN) and the elemental composition due to usage. The cutting fluid used in the machine tool is only analysed with respect to the elemental composition.

The hydraulic fluid is aged in a hydraulic circuit. In order to exactly simulate the loads applied to hydraulic fluids under operating conditions in reality the test stand is similar to industrially used stationary hydraulic systems. This means that a combination of different loads like temperature, oxygen as well as pressure are put on the fluid. Besides, the hydraulic circuit contains commonly used parts of typical industrially used hydraulic systems like a cooler, a tank and a filtration system. The flow circuit works as follows: the pump is working against a pressure relief valve which leads to a pressure of 35 MPa. Behind the pressure relief valve temperature is about 120 °C. The fluid flow into the tank is 45 l/min. Passing a cooler afterwards the fluid arrives at the suction side of the pump, again. Blank samples of the fluid are collected immediately after filling the test bench and mixing ("base fluid blank EN1-0" and "with additives blank EN1-0").

Two experiments with a plain bearing test bench are performed to evaluate the combination of fluid and materials. The hydraulic fluid is used with bronze as slide bearing material (CuSn8, free of cadmium and arsenic). It flows into the tank with 3 l/min. Passing a filter afterwards the fluid arrives at the plain bearing again. The tank is heated to a temperature of 80 °C (AGL-1 Table 1) or 90 °C (AGL-2, Table 1) in the bearing. The pressure in the bearing is adjusted to 2.5 N/mm^2.

The cutting fluid is used in a machine tool. It belongs to the group of metalworking fluids and its general task is cooling as well as lubrication during cutting operation. It is responsible for the following aspects: accelerated

Table 1
Characterisation of the tested fluids

	Parameter of usage	Kin. viscosity at 40 °C (mm^2/s)	Total acid number (mg KOH/g)
Cutting fluids			
Base fluid	Unused	7.7	0.2
Base fluid used ZDR	5 h, material: X5CrNi18-10, tank volume: 200 l	n.d.	n.d.
With additives	Unused	7.7	3.0
With additives blank ZDR-0	0 h, material: X5CrNi18-10, tank volume: 200 l	n.d.	n.d.
With additives used ZDR-1	30 h, material: X5CrNi18-10, tank volume: 200 l	n.d.	n.d.
Hydraulic fluids			
Base fluid	Unused	31.0	0.83
Base fluid blank EN1-0	0.5 h, 90–120 °C, 2.5 kW/l, 35 MPa, open system	31.0	0.84
Base fluid used EN1-1	308 h, 90–120 °C, 2.5 kW/l, 35 MPa, open system	459	31
With additives	Unused	31.9	1.0
With additives blank EN1-0	2 h, 90–120 °C, 2.5 kW/l, 35 MPa, open system	32.3	1.2
With additives used EN1-1	308 h, 90–120 °C, 2.5 kW/l, 35 MPa, open system	91.5	11.4
With additives used EN1-2	312 h, 90–120 °C, 2.5 kW/l, 35 MPa, closed system	32.1	1.2
With additives used AGL-1	80 °C, 1022 h, material: CuSn8	37	2.4
With additives used AGL-2	90 °C, 832 h, material: CuSn8	59.3	4.5
Mineral oil	Unused	35.6	2.0
Mineral oil used EN1-1	312 h, 90–120 °C, 2.5 kW/l, 35 MPa, open system	36.8	1.7

n.d. = not determined, ZDR = machine tool: cutting, EN1 = hydraulic test bench, AGL = plain bearing test bench.

heat dissipation with increased tool service life, lubrication between tool, workpiece and chips and improved chip removal. Fluids are analysed after 5 and 30 h of cutting operation with steel X5CrNi18–10 as work piece material and a tank volume of 200 l. A blank sample of the fluid with additives is collected immediately after filling the machine tool and mixing (“with additives blank ZDR-0”).

2.3. Viscosity and total acid number

Kinematic viscosity is a measure of the relative flow of a fluid under influence of gravity. The laboratory determination of viscosity has been carried out by using capillary tubes according to DIN 51562 (1978). The measurement is performed with a calibrated glass capillary viscometer, where the fluid passes through a fixed diameter orifice under the influence of gravity.

The TAN is measured according to DIN 51558 (1979). This method determines acidic and basic constituents in petroleum products.

2.4. Combined biodegradability test system

The batch assay called “O_2/CO_2 Headspace Test with GC-TCD” is performed following standard procedures ISO 10708 (BODIS-Test) and ISO 14593 (ISO CO_2 Headspace Test) with modifications. It is based on the measurement of the ultimate aerobic mineralisation of test substances to carbon dioxide in water. Results are expressed as biological oxygen demand in relation to the theoretical oxygen demand (BOD/ThOD) and as carbon dioxide production in relation to the theoretical carbon dioxide production (TIC/ThIC). The theoretical values are calculated on the basis of values from elemental analysis (Elemental Analyser Vario EL) in duplicate. The test substance is evaluated as “good (ready) biodegradable”, if degradation exceeds 60% within 28 days.

Test conditions are: 1 l infusion bottles closed with chlorobutyl stoppers (both: Zscheile & Klinger, Hamburg, Germany) and additional chlorobutyl septa (Macherey-Nagel, Dueren, Germany, N35B red); 800 ml mineral medium (ingredients as described in ISO 10708

Table 2
Soil characterization of LUFA 2.2

Parameter	Unit	Value
Kind of soil		Loamy sand[a]
Granulation size	(mm)	$\leqslant 2$[a]
pH-value		5.8 ± 0.2[a]
TOC	(%)	2.19 ± 0.08[a]
Water content	(%)	13.2
Dry weight	(%)	86.8
WHC_{max}	(g/100 g dw)	50 ± 4[a]
Cfu (fungi)	(cfu/g dw)	1.1×10^6
Cfu (bacteria)	(cfu/g dw)	9.5×10^6
Water extractable nitrite	(mg NO_2^-/kg dw)	<1.0
Water extractable nitrate	(mg NO_3^-/kg dw)	3.3
Water extractable phosphate	(mg PO_4^{3-}/kg dw)	0.6
Water extractable ammonium	(mg NH_4^+/kg dw)	0.1

[a] Specification LUFA, Speyer, Germany.

(1997)); test substances (100 mg ThOD) are directly weighed in; stirring (600 1/min); room temperature (20–25 °C). Standard soil Lufa 2.2 (Landwirtschaftliche Untersuchungs- und Forschungsanstalt Speyer, Germany) is used as the inoculum instead of activated sludge. The soil characteristics are described in Table 2. 2 g soil/l correspond to $\approx 10^7$ cfu of bacteria and fungi per liter medium.

Total carbon dioxide production is determined after acidification of liquid aliquots with 0.1 M orthophosphoric acid (1 ml) at the end of incubation in duplicate. The measurements are performed in 7 ml test tubes closed with GL 14 hole-caps and silicone septa (Alltech GmbH, Unterhaching, Germany, blue septa 1/2″). The sample volume is 2 ml using a gastight syringe (SGE, Austin, Texas, USA, Type 2.5MDR-GT with valve SGE, Type VLL25/2.5). The recovery of CO_2 is between 95% and 105%.

Oxygen and carbon dioxide in headspace samples are measured gas-chromatographically using a thermal conductivity detector (Perkin Elmer Autosystems) and a CTR 1 column (Alltech GmbH, Unterhaching, Germany). The sample volume of 100 μl is taken with a gastight syringe (Hamilton, Reno, Nevada, USA, Type 1710 SL). The analysis parameters are: oven temperature 40 °C isothermal, detector temperature 140 °C and gas flow helium (purity grade 99.999 vol.%) 62.5 ml/min. The single measurements are taken at least weekly. Calibration is done with three test gases (Linde AG, Duesseldorf) of the following compositions (vol.%): test gas 1: CO_2 (0.02), O_2 (20.8), N_2 (79.18); test gas 2: CO_2 (0.8), O_2 (20.8), N_2 (78.4); test gas 3: CO_2 (5.0), O_2 (5.0), N_2 (5.0), He (85.0).

Oxygen ($\approx$100 hPa) is redosed with a 100 ml gastight syringe (SGE, Austin, Texas, USA, Type 100MR-VLLMA-GT), if the oxygen partial pressure in the incubation bottles falls below 100 hPa. This is performed once within 28 days for good biodegradable substances.

Test substances, reference (sodium benzoate) and blanks are tested in triplicate. The replicability of degradation is indicated as standard deviation taking into account the standard deviations of the blank-values and of the biological oxygen demand or carbon dioxide production and an element of uncertainty of the ThOD or ThIC. The assumed elements of uncertainty are: 0.1 g O_2/g for the ThOD, 0.01 g CO_2/g for the ThIC, and 0.01 g O_2/g for single substances, where the values are calculated from the molecular formula.

2.5. Elemental analysis

The extent of biodegradation is expressed as a percentage of the theoretical O_2 uptake (ThOD) or CO_2 production (ThIC) for complete biodegradation of the test substance. These maximum values are normally calculated from the molecular formula. This is impracticable for most lubricants, because of their unknown chemical composition.

Elemental analysis is a usable method to calculate both ThOD and ThIC (Gerike, 1984 and Battersby, 2000), thus the theoretical values are calculated on the basis of values from elemental analysis (Elemental Analyser Vario EL) in duplicates. Furthermore, it allows to demonstrate oxidative changes of the lubricants.

3. Results

3.1. Viscosity and total acid number

The measure of internal friction in a fluid is viscosity. It is one of the most important properties for characterisation of lubricants and their flow and transport properties. The acidity of unused oils and fluids is normally derived from the type and concentration of the ester and specific additive materials, whereas the acidity of used oils is of interest to measure the degree of oxidation of the fluid. Both viscosity and TAN have been chosen to describe the degree of aging of the hydraulic fluids. The environmentally acceptable ester based oil was used in the hydraulic system as well as in the bearing test stand, whereas the mineral oil was only used in the hydraulic system. The mineral oil was only tested in a formulation including additives. The ester based oil was tested on the one hand as a base oil with no additives and on the other hand as a formulation incorporating additives. Viscosity and TAN of the used fluids are presented in Table 1. First of all, it can be seen that oxidation stability of mineral oil significantly exceeds the one of ester oils. Further, investigations show that test runs based on exposing the fluid to oxygen age the fluids the most. Finally, results from plain bearing tests show the strong influence of temperature on the oxidation process (hydraulic fluids with additives used AGL-1 and AGL-2).

3.2. Elemental analysis

The results of elemental analysis for the tested lubricants are shown in Table 3. The elemental compositions of all cutting fluids are equal except for sample “with additives used ZDR-1” with a slightly decreased hydrogen content. No significant effects of usage or additives can be detected.

The same is true for the hydraulic fluids with two exceptions: the hydraulic fluids “base fluid used EN1-1” and “with additives used EN1-1” show an increase of oxygen concentration of 12.2% up to 18.1% for the base fluid and of 12.5% up to 15.5% for the fluid with additives caused by the thermic-oxidative operating conditions. Taking into account also the increase of viscosity and the lower solubility in pentane, it can be concluded, that both fluids were partly oxopolymerised. The low oxygen concentrations of the mineral oils are probably caused by the additives. Due to the high oxygen content the ester oils have lower ThOD and ThIC values than the mineral oils.

3.3. Biodegradability

The natural standard soil Lufa 2.2 is used as inoculum in the described biodegradation test system. Other soil samples with similar characteristics can also be used. The concentration of aerobic and facultatively anaerobic bacteria and fungi should be in the range of 10^6– 10^8 cfu/l. The oxygen consumption in the blanks should not exceed 10 mg/l within 28 days (10% of the ThOD). For this reason 2 g soil/l are used as inoculum in this work.

Biodegradation kinetics of the base fluids and the reference substance sodium benzoate are shown in Fig. 1. The less distinct first order kinetic of the hydraulic base fluid can be explained by solubility and distribution characteristics of the substance, which causes transport limitations and may cause linear degradation kinetics. In these cases, the maximum rate of biodegradation is limited and a pass within the 10-day window is unlikely, because the saturation constant of the bacteria is high in relation to solubility. So this criterion for “ready” biodegradability is inappropriate, and in many cases incubation may need to be continued beyond 28 days before biodegradation reaches a plateau (Battersby, 2000). Therefore, the 10-day window criterion is not applied in the O_2/CO_2 Headspace Test with GC-TCD and only the degradability after 28 days is applied under qualitative consideration of the kinetics. In contrast to the hydraulic base fluid, the cutting base fluid shows an increase of biodegradation velocity with a similar

Table 3
ThOD and ThIC of the fluids calculated via elemental analysis (CHN-analyser)

	C (%)	H (%)	O (%)[a]	ThOD (g O_2/g)	ThIC (g CO_2/g)
Cutting fluids					
Base fluid	72.9	12.4	14.6	2.80	2.67
Base fluid used ZDR	72.8	12.1	15.1	2.76	2.67
With additives	72.8	12.3	14.9	2.78	2.67
With additives blank ZDR-0	72.5	12.0	15.5	2.74	2.66
With additives used ZDR-1	72.9	10.9	16.3	2.65	2.67
Hydraulic fluids					
Base fluid	75.9	11.2	12.9	2.79	2.78
Base fluid blank EN1-0	76.1	11.8	12.2	2.85	2.79
Base fluid used EN1-1	70.9	10.9	18.1	2.59	2.60
With additives	75.9	11.8	12.2	2.85	2.78
With additives blank EN1-0	75.2	12.3	12.5	2.87	2.75
With additives used EN1-1	72.6	11.9	15.5	2.73	2.66
With additives used EN1-2	76.4	12.0	11.6	2.88	2.80
With additives used AGL-1	75.8	11.6	12.6	2.83	2.78
With additives used AGL-2	74.8	11.5	13.7	2.78	2.74
Mineral oil	85.0	13.6	1.4	3.35	3.11
Mineral oil used EN1-1	84.9	13.4	1.6	3.32	3.11

ZDR = machine tool: cutting, EN1 = hydraulic test bench, AGL = plain bearing test bench.

[a] O (%) = 100 − (H (%) + C (%)).

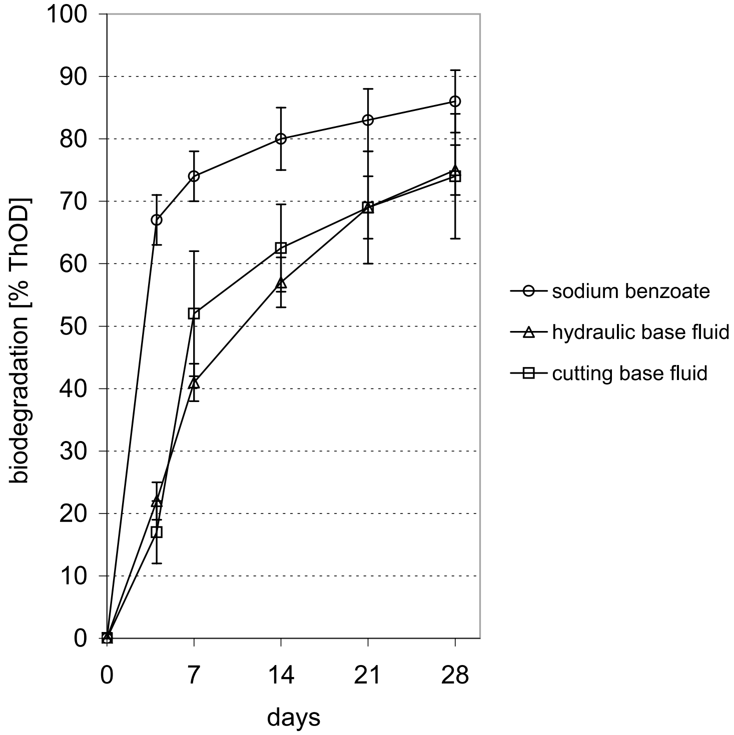

Fig. 1. Biodegradation kinetics of sodium benzoate, hydraulic fluids and cutting base fluids; standard deviation of three parallels.

lag-phase. The 10-day window criterion is passed, although water solubility is low.

During the cutting process the lubricants are exposed to high temperatures and pressures. Despite of these extreme conditions, no effects on biodegradability can be detected for the "base fluid used ZDR" as well as for the formulated fluid "with additives used ZDR-1" (Fig. 2). This can be explained both by the large oil volume of 200 l at this machine tool and by evaporation of used lubricants during operation. Therefore, only slight

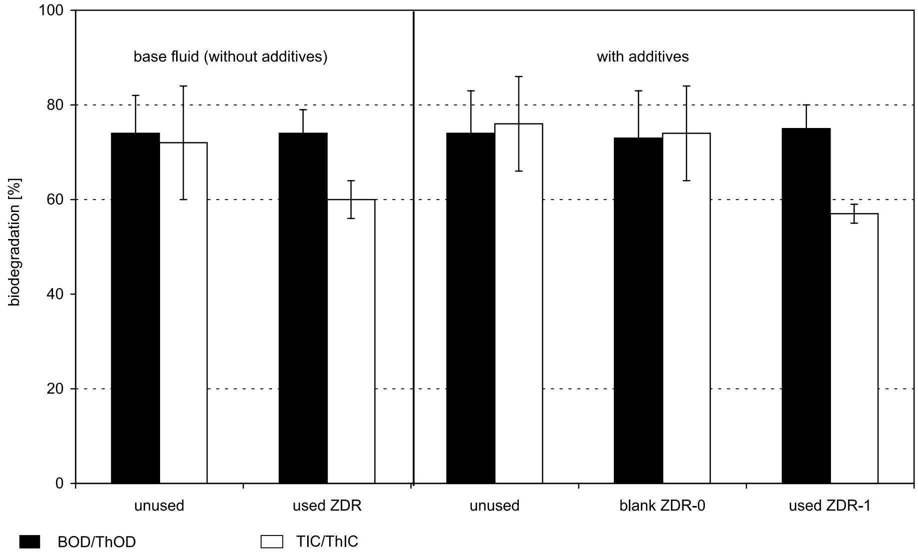

Fig. 2. Biodegradability of cutting fluids; ZDR = machine tool with cutting operations; standard deviation of three parallels.

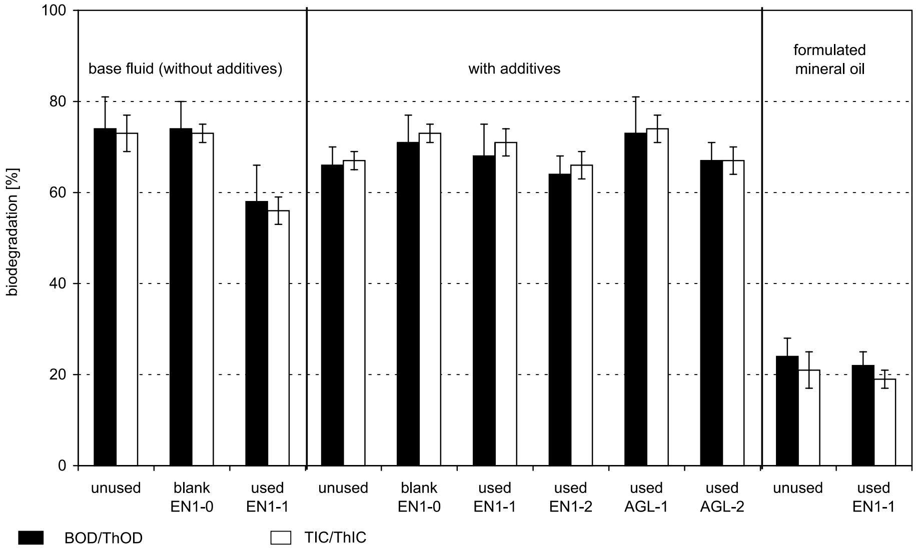

Fig. 3. Biodegradability of hydraulic fluids; EN1 = hydraulic test bench, -0 = blank, -1 = open system, -2 = closed system; AGL = plain bearing test bench, -1 = 80 °C, -2 = 90 °C; standard deviation of three parallels.

changes occur. Furthermore, no effects of additives on biodegradation can be shown.

The additives also have no respectively a slight influence on the biodegradability of the hydraulic fluids (Fig. 3). Biodegradability of the oxopolymerised "base fluid used EN1-1" is lower than the biodegradability of the "unused base fluid" and the "base fluid blank EN1-0". It even is below the OECD limit value of 60% for ready biodegradability. In contrast to this, no usage effects can be detected in the hydraulic fluids with additives.

As expected, mineral oils are far less biodegradable than the ester oils and their biodegradability is not affected significantly by usage.

4. Discussion

Most oleochemical esters as base fluids fulfill the requirements of the eco-labels. However, the additives necessary to achieve the required technical performance are often the limiting components (Willing, 1999). Randles (1992) showed, that additives can have a significant positive or negative effect on biodegradation of esters, applying the CEC-test. For the fluids presented here, the influence of additives on biodegradability is not significant. Like described by Battersby (2000) it is only depending on the kind of base fluids. The theoretical values of O_2 uptake and CO_2 production proposed by Battersby (2000) are confirmed by the results obtained with the mineral oils (ThOD = 3.4, ThIC = 3.1). In contrast to biodegradation tests, water accommodated fractions are applied for toxicity testing. Bennett et al. (1990) showed that mineral oil toxicity is depending only on the additives and not on the base fluids.

In this work the fluids were tested under extreme operating conditions at machine tools and test benches. For example the "hydraulic fluid" is exposed to high temperature (90–120 °C), pressure (35 MPa) and shearing forces. For the investigation of thermic-oxidative aging processes this test bench can be operated openly or closed against oxygen. Despite of these experimental conditions no effects can be detected except for one sample without additives: biodegradability in this case was below 60% within 28 days. Like the increase of viscosity and the CHN-analysis showed, the fluid was polymerised oxidatively. Such polymerisations could also be observed for hydraulic oils based on rapeseed oils (Zeman, 1997). Therefore, oxidative inhibitors are necessary as additives to prevent these changes. In contrast, Remmele and Widmann (1998) assumed, that there is no explicit relation between polymerisation with branched carbon chains and lower biodegradability.

Necessary antioxidant concentrations can be reduced by changing the base fluid esters chemically without reducing performance characteristics of the oils. By these means, risks caused by toxic effects can be reduced, too. A promising approach is followed at the "Institute of Technical Chemistry and Heterogene Catalyses" at the RWTH Aachen. The double bindings of the fatty acids responsible for polymerisation are saturated with carbon acids. This results in an increase of oxidation stability and nearly unchanged filterability at low temperatures (Keller et al., 2000).

5. Conclusion

The combined batch assay "O_2/CO_2 Headspace Test with GC-TCD" is well suited for the testing of biodegradability of lubricants. In this work, the effects of additives and usage are demonstrated. The additives in usual concentrations have no significant effect on the biodegradation of synthetic and native ester lubricants. This means, that biodegradability is depending mainly on the characteristics of the base fluids. Nevertheless, polymerisation effects may occur and reduce biodegradability, if the specific additives preventing these effects are missing or if their concentration is lowered during usage. Further effects influencing biodegradability have not been observed in this work.

For these reasons antioxidants like phenolic or aminic substances are necessary to prevent oxopolymerisation. These substances are often environmentally harmful and ecotoxic, and their concentration should be lowered by a simultaneous increase of oxidation stability of the base fluids. Lubricants are emitted both into the aquatic and the terrestric environment. Therefore, biodegradability should also be assessed in soil.

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