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Comparison of mixed trialkyl phosphine oxides (TRPO) extractants from different sources

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Abstract

Mixed trialkyl phosphine oxides (TRPO) are promising extractants. They may be used to recover radionuclides from highly radioactive liquid wastes. However, since TRPOs contain alkyl chains ranging from C4 to C8, different TRPO may have different characteristics. In this paper, four TRPOs from different sources are compared. The extraction and back-extraction characteristics for UO_2^{2+}, Pu^{4+}, Am^{3+}, and TcO_4^- were investigated, and the loading capacities for both of UO_2^{2+} and HNO_3 were compared. Infrared spectroscopy and pyrolysis gas chromatography were used to analyze the type and composition of the side-group in these TRPOs. The experimental results demonstrate that different TRPOs possess similar extraction and back-extraction characteristics because they have the same type of side-group. However, different TRPOs show different loading capacity for UO_2^{2+} and HNO_3 due to differences in the alkyl composition of the side-group.

Keywords: TRPO; Extraction; Back-extraction; Loading capacity; Side-group

1. Introduction

Disposal of highly radioactive liquid wastes produced in the nuclear power industry is a major factor that has limited the growth of nuclear power as an alternative source of energy. At the present time, there are two main approaches for disposal of highly radioactive liquid wastes: solidification and chemical partitioning. Solidification has been used commercially but its drawback is that a high level of radioactivity will remain for millions of years due to the presence of long half-life nuclides. Further, if some of these nuclides could be recovered, they could be reprocessed and reused. Chemical partitioning has not been used commercially although research has lead to the development of a number of processes, such as TRUEX [1–5], TRPO [6–9], DIDPA [10,11], DIAMEX [12], and CTH [13]. The TRPO process developed by Zhu et al. [8] and Zhu [9] is regarded as one of the most promising for commercialization [14].

The main objective of the chemical partitioning process is to recover all of the radioactive nuclides including uranium, transuranium, technetium, and cesium so that the residual liquid waste has only a medium level of radioactivity. The key to this accomplishment is to find an appropriate extractant for extracting these nuclides. Mixed trialkyl phosphine oxides (TRPOs) have been found to be very good extractants for uranium, trivalent transuranium, as well as technetium. Compared to the pure trialkyl phosphine oxides such as trioctyl phosphine oxide (TOPO), TRPO not only has a high loading capacity for UO_2^{2+} and HNO_3, but also a lower manufacturing cost and is thereby more suitable for industrial application. However, because TRPO is a mixture of trialkyl phosphine oxides, TRPOs from different sources may have different side groups and thereby a different molecular compositions. This composition difference may lead to different physico-chemical characteristics such as extraction, back-extraction, and loading capacity. These differences could lead to operational difficulties if TRPOs are used commercially. In this paper, the physico-chemical characteristics of TRPOs from different source are compared.

2. Experimental

2.1. Regents

Four TRPOs were purchased from Jinan Phosphate Fertilizer Factory (Shandong, PR China), Guangzhou Pharmacy Factory (Guangdong, PR China), CYTEC (Stamford, CT, USA, known as SYANEX 923S), and Shanghai No.

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4 Reagent Factory (Shanghai, PR China). These four TRPOs are designated as TRPO(I), TRPO(II), TRPO(III) and TRPO(IV). Hydrogenated kerosene was purchased from Jinzhou Refinery Factory (Liaoning, PR China) and was pre-distilled to accumulate the 180–220 °C components before use. A 30% TRPO/kerosene mixture was prepared and pre-washed successively by nitric acid, sodium carbonate, nitric acid, and water before use.

$UO_2(NO_3)_2$ was obtained by dissolving analytical grade UO_2 in concentrated nitric acid. Cs and Am, tagged by ^{137}Cs and ^{241}Am, were purchased from the China Institute of Atomic Energy (CIAE, Beijing, PR China). Pu, tagged by ^{239}Pu, was made at the Institute of Nuclear Energy Technology (INET), Tsinghua University (Beijing, PR China). Tc, tagged by ^{99}Tc, was purchased from Great Britain. These nuclides were all of radiochemical grade. Other reagents were all of analytical or chemical grade.

2.2. *Experimental condition*

In all the experiments, the ratio of the organic to the aqueous phase was unity. The experimental temperature was $25 \pm 1\,^{\circ}C$, and the organic phase was pre-balanced with a nitric acid solution at the same concentration as the aqueous phase in the extraction experiments.

2.3. *Analytical methods*

The concentration of nitric acid was determined by titration using standard sodium hydroxide with oxalic acid—potassium oxalate as a complexing agent. UO_2^{2+} was analyzed by a calorimetric method. Pu^{4+}, TcO_4^-, Cs^+, and Am^{3+} were determined by liquid-scintillation counting.

3. Results and discussion

3.1. *Comparison of physical characteristics of TRPOs*

The main physical characteristics of the four different 30% TRPO/kerosene mixtures are listed in Table 1. It can be seen that there are no distinct differences in density, viscosity, surface tension and refractive index. Although the solubility of water in TRPOs does not show a large variation, the initial water content in the TRPOs does vary, perhaps due to different manufacturing process. In this paper, the total organic carbon (TOC) is used to characterize the solubility of TRPOs in water and nitric acid solutions. It was found that TRPO(I) and TRPO(II) have a greater solubility in water and nitric acid solution than TRPO(III) and TRPO(IV). This could be attributed to the presence of some soluble impurities in TRPO(I) and TRPO(II). Further evidence of the presence of impurities is the deep color of these two TRPOs whereas pure TRPOs are colorless. After washing with acid solution, alkali solution, and water sequentially to about pH 7, their solubilities in water are lowered, suggesting a decrease of the contaminants. In addition, these two TRPOs were less colored.

3.2. *Comparison of extraction characteristics of TRPOs*

3.2.1. *Extraction of* UO_2^{2+}

Fig. 1 shows the relationships between the distribution ratio of UO_2^{2+} and nitric acid concentration in the aqueous phase when UO_2^{2+} is extracted by four different 30% TRPO/kerosene mixtures. In all cases, the total uranium content was 5 g/l. It can be seen from the figure that the distribution ratios of UO_2^{2+} are very high (mostly are over 1000). This confirms that all TRPOs have a very strong extractability for UO_2^{2+}. In general, the distribution ratio of UO_2^{2+} decreases as the nitric acid concentration increases for the four extractants.

3.2.2. *Extraction of* Pu^{4+}

The variation of the distribution ratio of Pu^{4+} with nitric acid concentration is shown in Fig. 2. Under the experimental condition, the distribution ratios of Pu^{4+} for all four 30% TRPO/kerosene mixtures are higher than 100. This result demonstrates that all four TRPOs are very strong extractants for Pu^{4+}. When the nitric acid concentration is lower than 2 mol/l, the distribution ratio of Pu^{4+} decreases as the nitric acid concentration increases. For a nitric acid

Table 1
Main physical characteristics of the four different TRPOs

	Color	Density ($kg\,m^{-3}$)	Viscosity (Pa s)	Surface tension ($N\,m^{-1}$)	Water content (%)	Refractive index
TRPO(I)	Buff	789.8	2.856×10^{-3}	33.15×10^{-3}	1.162	1.4663
TRPO(II)	Brown yellow	787.8	2.690×10^{-3}	33.1×10^{-3}	0.432	1.4660
TRPO(III)	Colorless	787.8	2.819×10^{-3}	32.35×10^{-3}	2.329	1.4570
TRPO(IV)	Colorless	787.8	2.731×10^{-3}	32.7×10^{-3}	3.511	1.4618

	Solubility of water in TRPOs (%)	Solubility of TRPOs in water ($mg\,l^{-1}$)	Solubility of washed TRPOs in water ($mg\,l^{-1}$)	Solubility of TRPOs in nitric acid solution ($mg\,l^{-1}$)
TRPO(I)	9.243	250.2	70.4	163.3
TRPO(II)	9.382	289.7	122.1	273.1
TRPO(III)	8.541	33.1	30.2	19.5
TRPO(IV)	9.394	33.5	20.7	26.6

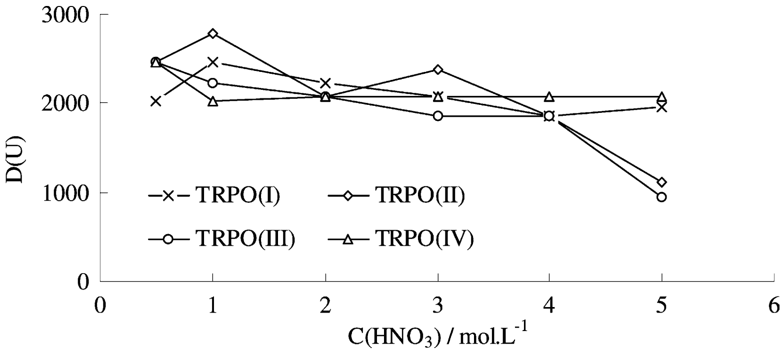


Fig. 1. Relationship between distribution ratio of UO_2^{2+} and nitric acid concentration in aqueous phase when UO_2^{2+} is extracted by four different 30% TRPO/kerosene mixtures.

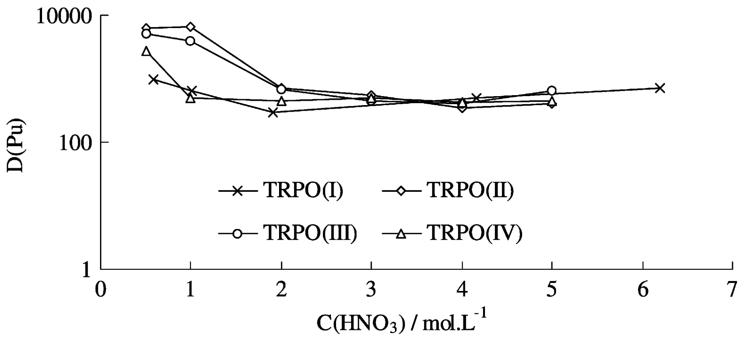


Fig. 2. Relationship between distribution ratio of Pu^{4+} and nitric acid concentration in aqueous phase for the four different 30% TRPO/kerosene mixtures.

concentration higher than 2 mol/l, the distribution ratio is nearly constant.

3.2.3. *Extraction of Am^{3+}*

The variation of the distribution ratio of Am^{3+} with nitric acid concentration in the aqueous phase is shown in Fig. 3. The four 30% TRPO/kerosene mixtures gave highly consistent results. As the nitric acid concentration increases, the distribution ratio of Am^{3+} decreases sharply. At a low nitric acid concentration, TRPOs have a high extractability for Am^{3+}. However, the distribution ratio decreases below unity when the nitric acid concentration is higher than 5 mol/l.

3.2.4. *Extraction of TcO_4^-*

The variation of the distribution ratio of TcO_4^- with nitric acid concentration is plotted in Fig. 4. As was the case of Am^{3+}, highly consistent results are attained for all four 30% TRPO/kerosene mixtures. As the nitric acid concentration increases, the distribution ratio of TcO_4^- decreases. However, unlike Am^{3+}, the distribution ratio of TcO_4^- is still around unity, even for nitric acid concentrations higher than 5 mol/l. Importantly, TcO_4^- and Am^{3+} can not be separated completely if the organic phase containing Am^{3+} and TcO_4^- is back-extracted with 5 mol/l nitric acid.

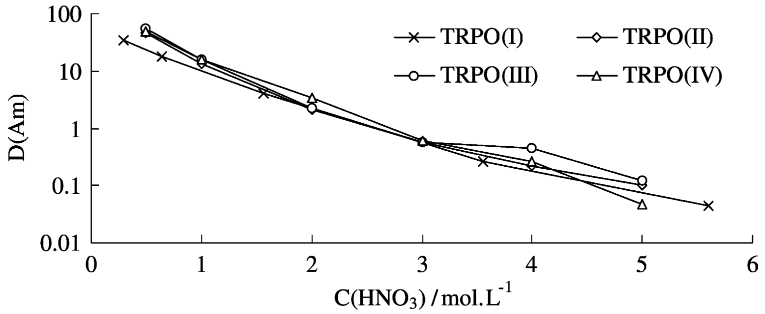


Fig. 3. Relationship between distribution ratio of Am^{3+} and nitric acid concentration in the aqueous phase when Am^{3+} is extracted by four different 30% TRPO/kerosene mixtures.

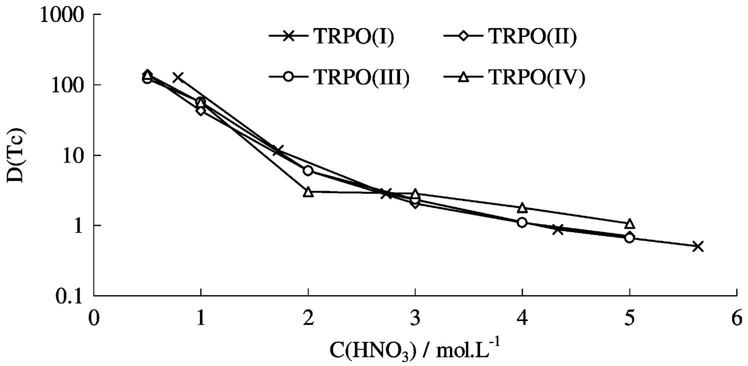


Fig. 4. Relationship between distribution ratio of TcO_4^- and nitric acid concentration in aqueous phase when TcO_4^- is extracted by four different 30% TRPO/kerosene mixtures.

3.2.5. *Extraction of Cs^+*

In Table 2, some extraction distribution ratios of Cs^+ with four 30% TRPO/kerosene mixtures are listed. It can be seen from the table that Cs^+ can not be extracted into the organic phase by any of the four TRPOs.

3.3. *Comparison of back-extraction characteristics of TRPOs*

3.3.1. *Back-extraction of Am^{3+} with high concentrations of nitric acid*

The effect of nitric acid concentration on the back-extraction ratio of Am^{3+} is shown in Table 3 for the four 30% TRPO/kerosene mixtures. It can be seen that as the nitric acid concentration increases, the back-extraction ratio increases. When the nitric acid concentration is higher than 5.0 mol/l, the back-extraction ratio of Am^{3+} for all four organic phases is higher than 89%. It may be concluded that the higher the concentration of nitric acid, the higher the efficiency for back-extraction of Am^{3+} for all four 30% TRPO/kerosene mixtures.

Table 2
Relationships between distribution ratio of Cs^+ and nitric acid concentration in the aqueous phase when Cs^+ was extracted with four 30% TRPO/kerosene mixtures

$C(HNO_3)$ ($mol\,l^{-1}$)	$D_{(Cs)}$			
	TRPO(I)	TRPO(II)	TRPO(III)	TRPO(IV)
0.5	3.80×10^{-4}	2.90×10^{-4}	9.60×10^{-5}	5.00×10^{-4}
1.0	1.87×10^{-4}	1.30×10^{-4}	4.10×10^{-4}	5.90×10^{-4}
2.0	1.00×10^{-4}	3.20×10^{-4}	2.90×10^{-4}	5.70×10^{-4}
3.0	1.00×10^{-4}	3.90×10^{-4}	1.80×10^{-4}	2.00×10^{-4}
4.0	$<10^{-4}$[a]			
5.0				

[a] The organic phase concentration of Cs^+ were lower than the analytical limit.

Table 3
Back-extraction ratio of Am^{3+} with nitric acid from four 30% TRPO/kerosene mixtures (%)

$C(HNO_3)$ ($mol\,l^{-1}$)	TRPO(I)	TRPO(II)	TRPO(III)	TRPO(IV)
3.0		64.4	63.8	62.5
3.6	79.4			
4.0		82.0	69.2	79.4
5.0		90.7	89.0	95.4
5.6	95.9			

3.3.2. *Back-extraction of Pu^{4+} with oxalic acid*

From Table 4, it can be seen that Pu^{4+} can be back-extracted efficiently with 0.6 mol/l oxalic acid from the four different 30% TRPO/kerosene mixtures. However, differences still exist. Among the four different organic phases, TRPO(I) shows the best characteristics for back-extraction of Pu^{4+} leading to 99.9% Pu^{4+} back-extraction into the aqueous phase with oxalic acid.

3.3.3. *Back-extraction of UO_2^{2+} with sodium carbonate*

The carbonate ion can form a strongly chelated compound. Thus, it is widely used to back-extract UO_2^{2+} from organic phases. In Table 5, the back-extraction ratio of UO_2^{2+} with 5% Na_2CO_3 for four different 30% TRPO/kerosene mixtures are compared. It can be seen that TRPO(III) has a lower back-extraction efficiency of UO_2^{2+} with sodium carbonate than the other three TRPOs.

Table 4
Back-extraction ratio of Pu^{4+} with oxalic acid from four different 30% TRPO/kerosene mixtures (%)

TRPO(I)	99.9
TRPO(II)	78.3
TRPO(III)	84.7
TRPO(IV)	78.6

Table 5
Back-extraction ratio of UO_2^{2+} with sodium carbonate from different 30% TRPO/kerosene mixtures (%)

TRPO(I)	92.2
TRPO(II)	93.2
TRPO(III)	81.7
TRPO(IV)	96.3

Table 6
Distribution ratio of TcO_4^- during the successive processes

	Concentration							H_2O
	HNO_3		$H_2C_2O_4$	$(NH_4)_2CO_3$				
	1.0 mol/l	5.0 mol/l	0.6 mol/l	5%	0.1%	0.1%	0.1%	
TRPO(I)	59.8	0.65	35.9	16.0	2.17	0.67	0.81	0.14
TRPO(II)	56.5	0.66	42.4	47.2	4.79	2.17	1.73	2.82
TRPO(III)	42.9	0.7	15.6	11.3	3.11	0.27	0.35	0.45
TRPO(IV)	55.4	1.07	48.5	33.8	3.26	0.98	1.91[a]	0.82

[a] The third phase appeared.

During the back-extraction of UO_2^{2+} with 5% $(NH_4)_2CO_3$ or Na_2CO_3 from TRPO/kerosene mixtures, emulsification can be observed. Emulsification may lead to operational difficulties and phase separation. When the experimental temperature was lower than 20 °C, except for TRPO(I), two clear phases could not be separated for the three other TRPOs even if these sample were left to stand for more than 30 min. However, centrifugation can improve phase separation. When the temperature was higher than 20 °C and the mixtures were left to stand for a long time, good phase separation occurred. This implies that the centrifugal extractor may be more suitable during the back-extraction of UO_2^{2+} from 30% TRPO/kerosene with carbonate ions.

3.3.4. Back-extraction of TcO_4^-

TcO_4^- is a very troublesome ion in the TRPO process because it co-back-extracts into several streams with other nuclides [15]. About 75% of TcO_4^- is back-extracted into the aqueous phase together with Am^{3+}/Cm^{3+} by a 5.5 mol/l nitric acid solution. About 6% TcO_4^- is back-extracted by the carbonate ion together with UO_2^{2+}. The remaining 18% TcO_4^- remains in the organic phase. In a previous study [15], it was shown that TcO_4^- remaining in the organic phase could be back-extracted efficiently with an alkali at about pH 8. However, from Table 6, it can be seen that different TRPOs have different back-extraction efficiencies for TcO_4^-. In the experiments conducted here, TcO_4^- was extracted by all four 30% TRPO/kerosene mixtures from a 1 mol/l nitric acid solution. Then the organic phases were washed with a 5.0 mol/l nitric acid solution, 0.6 mol/l oxalic acid solution, 5% ammonium carbonate solution, 0.1% ammonium carbonate solution (three times), followed by DI water (the above process is also the main procedure for the TRPO process). From Table 6, it can be seen that the first time the organic phases were washed by 0.1% $(NH_4)_2CO_3$ at pH $\approx$ 8, the back-extraction efficiencies were not good. In the second and third times, most of the TcO_4^- in 30% TRPO(III)/kerosene and 30% TRPO(I)/kerosene was back-extracted. In the successive water stripping process, most of the TcO_4^- also could be back-extracted except for the TRPO(II). The distribution ratio of TcO_4^- in TRPO(II) is greater than unity implying poor back-extraction with this method. However, from Fig. 4, it can be concluded that TcO_4^- can be back-extracted efficiently from all four TRPOs when using at least 7 mol/l nitric acid solution.

3.4. Comparison of loading capacities of TRPOs

3.4.1. Loading capacity of UO_2^{2+}

The loading capacity of UO_2^{2+} extracted by the four different 30% TRPO/kerosene mixtures is characterized by the loading organic capacity (LOC) value of UO_2^{2+}. The LOC value is defined as the highest concentration of UO_2^{2+} in the organic phase before the formation of a third phase. Fig. 5 shows the relationships between the LOC value of UO_2^{2+} and the nitric acid concentration in the aqueous phase at 30 °C for the four different 30% TRPO/kerosene mixtures. In all of the cases, the total concentration of UO_2^{2+} in the two phases is 100 g/l. It is apparent that a large difference in the LOC value of UO_2^{2+} exists among the

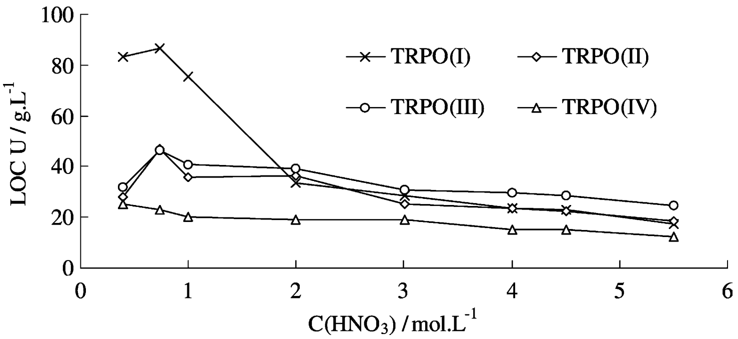


Fig. 5. Variations of LOC values of UO_2^{2+} with nitric acid concentration in the aqueous phase.

Table 7
Loading capacity of HNO_3 with four different 30% TRPO/kerosene mixtures (described by third phase disappearance temperature, °C)

$C(HNO_3)$ ($mol\,l^{-1}$)	TRPO(I)	TRPO(II)	TRPO(III)	TRPO(IV)
5.0	8	12	5	17
7.6	31	37.6	26.7	43
10.0	60.5	64	59.5	74
12.1				
14.7	>74 °C[a]			
15.8				

[a] Decomposition process of nitric acid begin.

four TRPOs. TRPO(II) and TRPO(III) have similar loading capacity for UO_2^{2+} and their LOC value for UO_2^{2+} is also consistent with that of TRPO(I) if the nitric acid concentration is higher than 2 mol/l. However, very high LOC values of UO_2^{2+} were obtained at low nitric acid concentrations (less than 2 mol/l) for TRPO(I). Over the entire range of nitric acid concentrations studied, the LOC values of UO_2^{2+} in TRPO(IV) are much lower than for the other three TRPOs.

3.4.2. Loading capacity of HNO_3

In Table 7, the loading capacity of nitric acid in the four 30% TRPO/kerosene mixtures is compared. The loading capacity of HNO_3 is given by the temperature at which the third phase disappears when the HNO_3 solution is contacted with the various 30% TRPO/kerosene mixtures at equal phase volume. It can be concluded from Table 7 that the loading capacity of the nitric acid in the four 30% TRPO/kerosene mixtures decreases in the order TRPO(III) > TRPO(I) > TRPO(II) > TRPO(IV). At nitric acid concentrations above 12 mol/l, the third phase does not disappear even at a temperature above 74 °C. However, the nitric acid begins to decompose.

3.5. Comparison of spectrums of TRPOs

Fig. 6 gives the infrared sorption spectra for the four TRPOs. It can be seen that the spectra of the different TRPOs are very similar. In these plots, the 2850–2950 cm^{-1} position denotes the C–H stretching vibration, the 1465 cm^{-1} position denotes the C–H asymmetric deformation vibration, the 1382 cm^{-1} position denotes the C–H symmetric deformation vibration, the 723 cm^{-1} position denotes the CH_2 phase rock vibration of $(CH_2)_n$ ($n > 4$), the 1155 cm^{-1} position denotes the P=O stretching vibration, and the 1405 cm^{-1} position denotes the P–C deformation vibration [16]. The two broad peaks at 3423 and 1640 cm^{-1} are not characteristic of the spectra of alkyl phosphine oxides. These two peeks, which are characteristic of hydroxyl groups, are a result of the presence of a small amounts of water and other alcohol impurities in the TRPOs.

It also can be seen that the spectra of TRPO(II) and TRPO(III) are more similar, especially in the position range of 667–1333 cm^{-1}. This zone is called the fingerprint zone. Fig. 7 shows the infrared sorption spectra of TRPO(II) diluted with different amounts of CCl_4. It can be seen that after dilution to 30 and 1%, the P=O stretching vibration peak moved to 1159.0 and 1168.7 cm^{-1}, respectively, from 1145.5 cm^{-1}. This demonstrates the existence of a strong intermolecular interaction among TRPO molecules in pure TRPOs. These kinds of interaction result in the P=O stretching vibration position being moved to a lower wave number. This interaction may come from the hydrogen bond between the O atoms in the P=O group and the H atom in the alkyl chain. The strong repulsive force from the three long

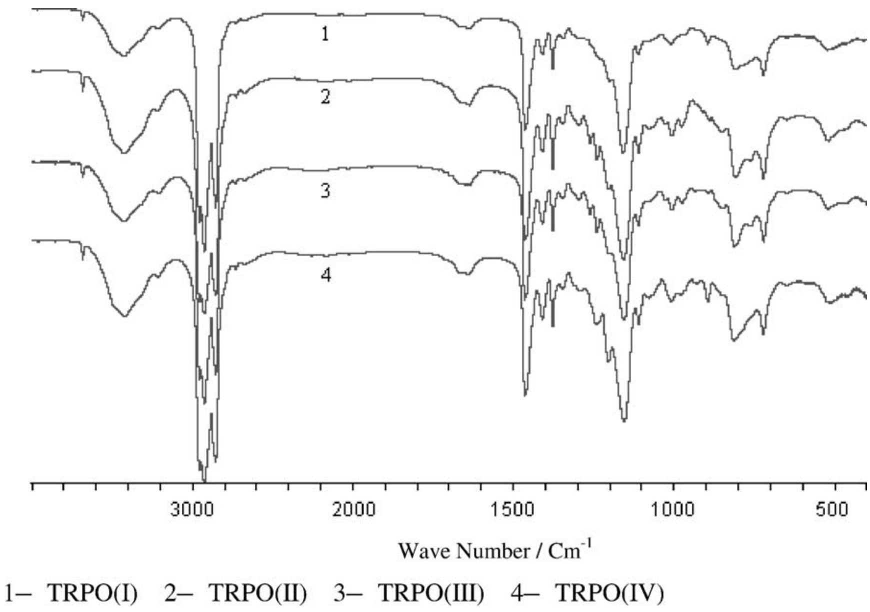


Fig. 6. Infrared sorption spectrograms for the four TRPOs.

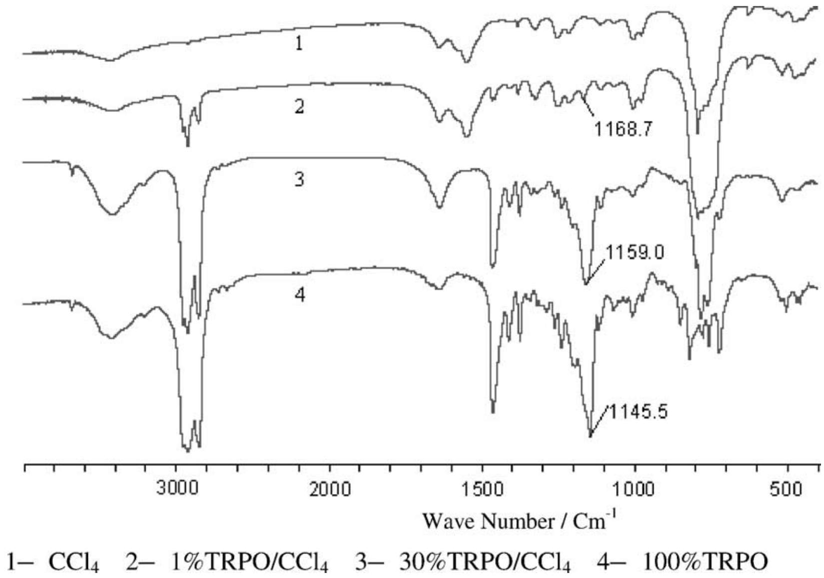


Fig. 7. Infrared sorption spectrograms of TRPO(II) diluted with different amounts of CCl_4.

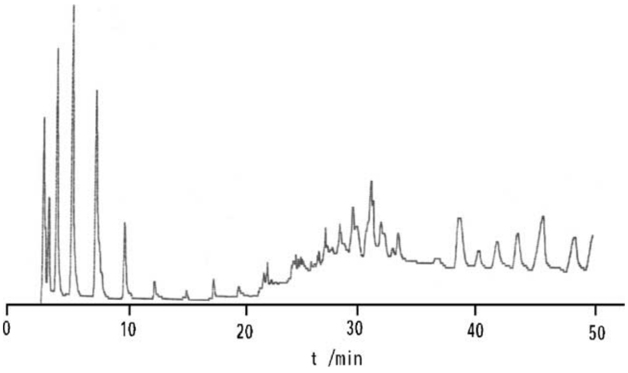
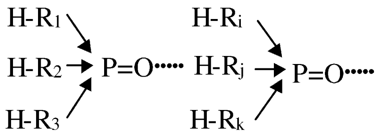


Fig. 8. Pyrolysis gas chromatography of TRPO(I).

alkyl chains makes the charge density around the O atom increase. Thus, the O atom can form a hydrogen bond with the hydrogen atom in alkyl chain as follow:



The addition of CCl_4 decrease the density of TRPO thus weakening the hydrogen bonds between two TRPO molecules. This results in the P=O stretching vibration peak moving to high wave numbers.

In Figs. 8–11, pyrolysis gas chromatography (PGC) analysis of the four TRPOs was carried out. PGC can break

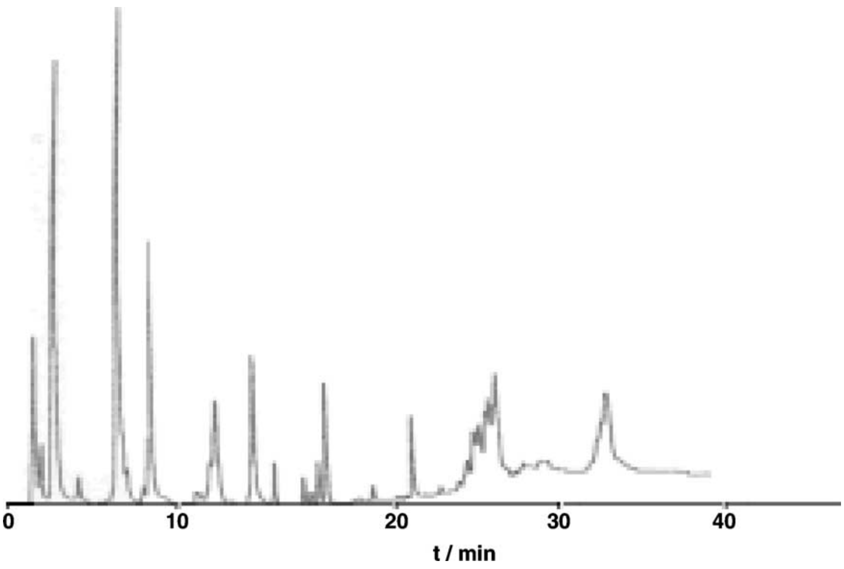


Fig. 9. Pyrolysis gas chromatography of TRPO(II).

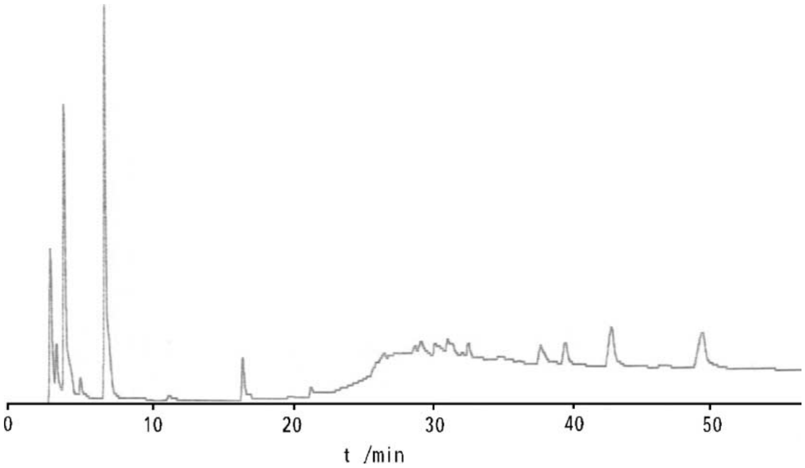


Fig. 10. Pyrolysis gas chromatography of TRPO(III).

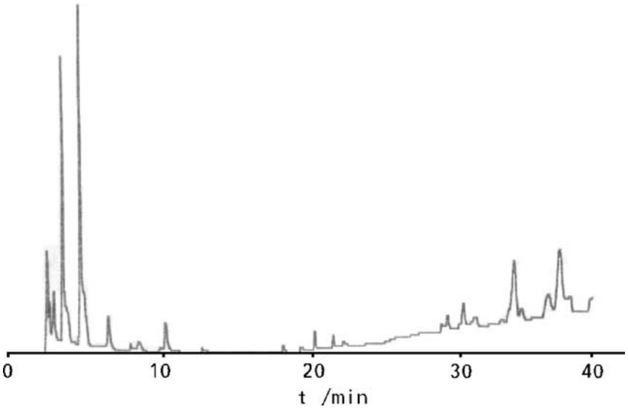


Fig. 11. Pyrolysis gas chromatography of TRPO(IV).

the P–C bond of TRPO. Then the type of alkyl side-group and content can be determined according to the peak position and the peak area [17]. As a standard spectrum, Fig. 12 shows the results of pyrolysis gas chromatography of trioctyl phosphine oxide (TOPO). From Fig. 12, it can be seen that TOPO only has one peak before 10 min (the other peaks are the decomposition compounds at successively higher temperatures). In the same period (before 10 min), TRPO(I) has six main peaks, TRPO(II) has four main peaks, and TRPO(III) has three main peaks. TRPO(IV) also has several peaks, however they appear earlier than the other TRPOs. This demonstrates that the alkyl compounds of TRPO(IV) are shorter than that of other TRPOs. From these spectra, it can be concluded that four TRPOs have different composition of alkyl side groups. TRPO(I) has the most complex composition. Since the four TRPOs have similar types of side-group, they showed similar extraction characteristics. However, because they have different side-group compositions, they showed different loading capacity. The more complex the side-group composition, the higher the loading capacity.

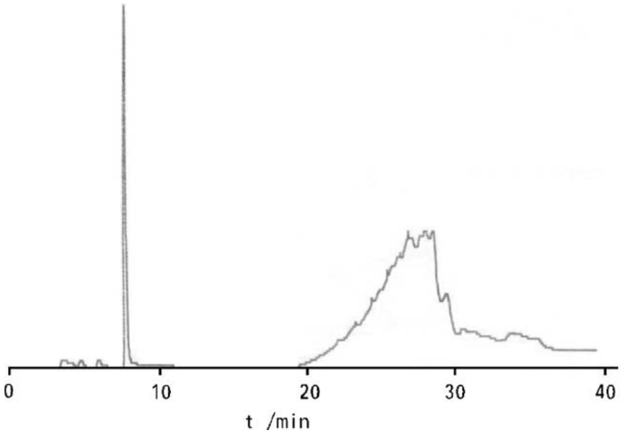


Fig. 12. Pyrolysis gas chromatography of TOPO.

4. Conclusion

Four different TRPOs were studied. The experimental results demonstrate that all four have similar physical characteristics except for color, water content and solubility in the aqueous phase. They also show similar extractive behavior for UO_2^{2+}, Pu^{4+}, Am^{3+}, TcO_4^-, and Cs^+. In the back-extraction experiments, the four TRPOs behave differently, especially for the back-extraction of TcO_4^- and UO_2^{2+}. The loading capacity of the four TRPOs also showed large differences, especially for the loading capacity of UO_2^{2+} at low concentrations of nitric acid. Infrared spectroscopy and pyrolysis gas chromatography analysis showed that the four TRPOs have the same types of side groups but different side-group compositions. Based on the experimental results, it can be concluded that different TRPOs have different physico-chemical characteristics. This implies that TRPO quality is an important factor if TRPO is to be used commercially.

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