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Application of magnetic fields to the decontamination and cleansing of Co-60 contaminated oils, through the extraction of ferromagnetic sludge

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Abstract

An inexpensive, alternative, simple technique to decontaminate and clean used-up oils slightly contaminated with Co-60, Mn-54 and Zn-65 is presented. This technique takes advantage of the ferromagnetic properties of the material suspended in the contaminated oils and is based on the use of small, commercial, inexpensive magnets. The preliminary results, which are also presented, demonstrate the feasibility of this technique when applied to oils at the laboratory scale. The efficiency of the technique makes possible an easy separation of the radioactive material in the form of sludge, which may be disposed of as any low-level radioactive waste. In accordance with Mexican normativity on radioactive wastes, the cleaned part of the oil is ready to be handled without the need of an incineration process.

1 Introduction

The operation of power nuclear reactors to generate electricity produces different types of radioactively contaminated materials [1,2]. One of these is obtained when the oils that lubricate engines, pumps and other mobile internal parts that contribute to the operation of the plant, get dirty and have to be changed. However, some chemical elements in the internals of the plant which are exposed to high neutron fluxes, are radio-activated. The internals release by several mechanisms, microscopic particles that are then incorporated into the lubricants. Among the radioactive elements which are generated in the process are Mn-54, Zn-65 Cs-137 and Co-60. The last one prevails longer due to its long mean life (5.27 years).

According with radiological safety regulations, the radioactively contaminated used-up oils must be treated as radioactive wastes for later processing. In general special incineration techniques are used into specifically prepared ovens for the processing of these oils. Nevertheless these techniques are sometimes rejected by environmentalist societies. Therefore the present authors have developed a radiologically-safe, simple and inexpensive technique at the laboratory scale, to radioactively decontaminate and clean this kind of oils. This technique takes advantage of the ferromagnetic properties of the material suspended [3] in the contaminated oils and is based on the use of small, commercial, inexpensive magnets each one of which can produce magnetic fields of strength of up to 1000 Gauss. Results of the applications of the technique as well as its efficiency show that it can be used at a higher scale. The activity concentrations of the oils are in the range of 1E-06 to 1E-04 μCi/ml (3.7 E-02 to 3.7 Bq/ml) and are contained in 200 liter metallic drums of standard dimensions. A brief description of the experiments performed to establish this technique follows.

2 Qualitative observations

As the oils have been stored in the drums for several months, a natural phenomenon of precipitation of microscopically large solid particles which incorporate radioactive material, has been taking place. At the same time, a structure of layers of different thickness, density and composition, including water, has been generated inside the drums. The result of those processes is the formation of a heavy sediment which constitutes a sludge composed by different contaminants, metallic particles and dirt, including radioactive material. The consequences of those phenomena are the increment of the counting rate of the oils from top to bottom of the drums. This is observed in all cases as well as the fact that the oil in the upper shell shows a very low activity when compared with that of the oil in the other layers.

3 Semi-quantitative observations

Three drums with the more frequent activities in the order of 10^{-5}μCi/ml and an additional one with a higher activity (10^{-4}μCi/ml), were chosen to analyze the oil activity behavior. To this end, specific activity measurements at different depths were performed. Four samples 50 ml each were taken at 5, 20, 50 and 70 cm depth from which Co-60 and Mn-54 were identified and the activity corresponding to the photopeaks determined. Table 1 shows the results of the measurements. It can be deduced from this table that a precipitation of radioactive material was produced, although in some layers the activity does not follow the

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rule of its increasing with depth. This "anomaly" may be explained in terms of the particular density and composition of those layers, which could facilitate the transit of radioactive material to lower levels. As the important fact to be considered here was the general increment of the activity with depth, no attempt was done to deeply explain this "anomaly". In general the values of activity reported in Table 1, clearly show that a precipitation of solid particles, including radioactive material, was indeed produced. It must be remarked that the predominant radionuclide in the samples is Co-60 which is a ferromagnetic material.

Table 1. Activities of samples at different depth in four drums.

Sample Number	Activity (Bq)	
	Co-60	Mn-54
99AR-092-1	4.4	Traces
99AR-092-2	3.7	-----
99AR-092-3	4.6	-----
99AR-092-4	4.9	Traces
99AR-079-1	17.0	Traces
99AR-079-2	19.2	Traces
99AR-079-3	16.2	Traces
99AR-079-4	15.1	Traces
99AR-050-1	30.0	-----
99AR-050-2	29.0	-----
99AR-050-3	32.7	-----
99AR-050-4	634.7	Traces
99AR-036-1	92.3	-----
99AR-036-2	94.0	-----
99AR-036-3	89.2	-----
99AR-036-4	90.5	-----

4 Hypothesis

As the oils were used to lubricate motors, pumps and other metallic mobile parts, it is to be expected that they bring appreciable amounts of small particles of metallic residues containing iron, nickel and cobalt, all of which have ferromagnetic properties. The concentration of those residues may be much higher than the concentration of Co-60. Taking this into consideration, it is possible to assume that the dirt and the metallic residues along with the Co-60 could form clusters of microscopically large size. Those clusters precipitate by gravity towards the bottom of the drums to form an oily sludge. Starting from this assumption and from the measurements of the oil activity along the vertical dimension of the drums and taking advantage of the ferromagnetic properties of the contaminants, a magnetic method was devised to attract the clusters, faster than gravity, to the bottom of non-magnetic containers.

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5 First magnetic experiment

From the above discussion it is suggested that the gravitational precipitation could be accelerated by means of magnetic fields using commercial, inexpensive permanent magnets. Therefore a bed of those magnets was constructed in such a way that a field intensity of approximately 450 Gauss was obtained.

Two oil samples 500 ml each labeled "A" and "B" were taken from a drum number 99AR-050 at 50 and 75 cm depth, respectively. This drum was chosen due its high Co-60 activity, as it is shown in Table 1. The samples in glass containers were placed on the 450 Gauss magnetic bed in two sessions three days each, as explained below:

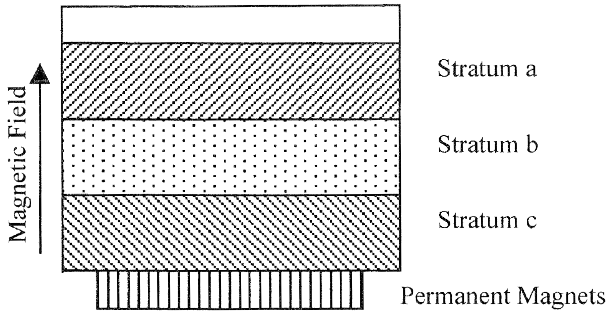

Figure 1. Strata structure after the application of the magnetic field. By mechanical filtering of oil from stratum "a", a portion denominated stratum "d" is obtained.

1. Sample "A" is submitted to the action of the magnetic field while sample "B" stays as a witness or control sample. No changes in the witness were detected.
2. After 3 days, Sample "A" is withdrawn from the magnetic field while Sample "B" stays as a witness. No changes in the witness were detected.
   It should be noticed that the results obtained in the first test with the sample "A" were not altered during the second test. Furthermore, it was observed that the samples "A" and "B" showed stratification in three caps during the process.
3. Next, 50 ml of oil from each of the three strata were taken out of samples "A" and "B" to be deposited respectively into six flasks and were analysed. Additionally, from both samples "A" and "B", filtering the oil extracted from the strata "a" with a fibber glass mesh generated a fourth sample. These two filtered samples were designated as stratum "d". Results are shown in Table 2.

Table 2. The activity are distributed in four strata

SAMPLE "A"			
Stratum	Description	Co-60 Photopeak 1173.24 keV	Co-60 Photopeak 1332.5 keV
1a	Magnetic cleaned oil	2755±61	2352±52
1b	Sludge	6909±93	6212±84
1c	Water	1557±10	1281±17
1d	Magnetic and mechanic cleaned oil	1973±11	1603±19
Sample "B"			
2a	Magnetic cleaned oil	2555±13	2125±23
2b	Sludge	9303±33	7003±41
2c	Water	703±7	526±13
2d	Magnetic and mechanic cleaned oil	1465±10	1295±16

Table 2 shows how the counts numbers for two photopeaks of Co-60 are distributed in the four strata of samples "A" and "B". The first stratum "a" contains clean oil free from the "organic-metallic granules". The oil cleaning process here was accomplished by the magnetic drag of the "granules" towards the second stratum "b", in which they have been accumulated to form sludge. This stratum floats on stratum "c". This last is composed mainly by moderately transparent water.

It must be emphasized at this point that the ratio of the activities corresponding to the "d" and "b" strata is greater than 6 for sample "B". This fact confirms the hypothesis used to try inexpensive magnetic means to clean oils contaminated with Co-60.

6 Second experiment

Before starting this second magnetic experiment, a 500 ml sample was obtained at a 10 cm depth from the 99AR-097 drum, in order to confirm that the specific activity in the upper layer is rather small as a consequence of natural precipitation. Results of measurements are reported in Table 3.

In this experiment the effects of the application of magnetic fields to a larger volume of contaminated oil were studied. For this, a cylindrical glass container 35 cm diameter and 40 cm height was used. A bed of several permanent magnets was constructed which produced an almost uniform 450 Gauss magnetic field parallel to the cylinder axis, at the bottom of the recipient.

5,000 ml of oil from drum 99AR-097 were taken from a 80 cm depth, then 500 ml of oil were taken from this volume and poured into a Marinelli type of

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recipient for activity measurement. The remaining 4,500 ml were magnetically treated. Another 5,000 ml sample was taken from this drum and left as a control sample in a cylindrical glass container like the one described above. The Marinelli recipient containing the 500 ml of the oil was introduced in a GeHp gamma spectrometry system for counting during 75,000 s, to obtain the spectrum and calculate the specific activity. The specific activity of this sample was 1.648 Bq/ml.

After three days of magnetic treatment, two 500 ml samples were obtained: one from the upper part and the other form the middle part of the cylindrical recipient container. Both samples were mechanically filtered, counted during 75,000 s and their specific activity determined. Finally 500 ml of oil were taken from the lower part of the recipient, then poured into a "Marinelli" container for counting during 75,000 s. This operation permitted the gathering of solid residues deposited in the cylindrical recipient bottom. Finally the specific activity was determined. Table 3 condenses the obtained results.

Data from Table 3 show that the application of the magnetic method produces a substantial decrease of the original activity of the oil and that the contamination is concentrated in the bottom of the recipient in the form of sludge. Table 3 shows also that the natural precipitation of contaminants reduces drastically the specific activity in the upper layers of the oil.

Table 3. Specific activities in strata a, b, c and g* of drum 99AR-097.

Stratum	Description (sample position)	Co-60 Photopeak 1173.24 keV Bq/ml	Co-60 Photopeak 1332.5 keV Bq/ml
a	High stratum	0.3176	0.3169
b	Middle stratum	0.3240	0.3250
c	Low stratum	2.4460	2.4663
g*	10 cm depth	0.0900	0.0940

*This sample was tacked to 10 cm depth, see text.

7 Third experiment

Drum 99AR-050 is characterised by a high specific activity and a high concentration of suspended organic-metallic contaminants. For this experiment a 5,000 ml sample of oil was taken from this drum at a 50 cm depth and poured into a cylindrical container like the one described before. From this amount, a first sample 500 ml was taken and poured into a Marinelli for counting during 75,000 s. From the analysis of the gamma spectrum the following values of the specific activity were found: 6.9647 Bq/ml for Co-60 and 0.0254 Bq/ml for Cs-137.

The remaining 4,500 ml were magnetically treated during three days. Afterwards two 500 ml samples were obtained: one from the upper part (a) and other from the lower part (c) of the cylindrical container; this one contains the sludge that was accumulated during the treatment. Oil of the upper part was mechanically filtered, counted during 75,000 s. and its specific activity quantified. Finally 500 ml of oil of the lower part of the cylindrical container was counted during 75,000 s and its specific activity quantified. Table 4 condenses the obtained results. It is seen from the above that the present magnetic cleaning technique can also be applied to oils of higher degree of contamination.

Table 4. Specific activities in high and low strata of drum 99AR-050.

Stratum	Description (Sample position)	^{137}Cs Photopeak 661.64 keV Bq/ml	Co-60 Photopeak 1173.24 keV Bq/ml	Co-60 Photopeak 1332.5 keV Bq/ml
a	High stratum	---------------	1.1249	1.1731
c	Low stratum	0.1054	18.2745	19.9324

Finally it must be mentioned that extraction of oil samples from drums was performed with the aid of a laboratory-made electronic probe based on an small inexpensive optically-coupled device and a common voltmeter. By monitoring the output voltage changes of the probe, the exploration through the layers of the oil in the drums was successfully done.

8 Conclusions

Used-up oils contaminated with Co-60, Mn-54, Zn-65 and Cs-137 have been storage during several months. Natural precipitation of contaminants and stratification of material took place in the drums, in such a way that an increment of the specific activity from surface to bottom was observed. Measurements of the specific activity of material in the different layers confirmed this fact. As the oils were used to lubricate mobile metallic parts, microscopically large metallic particles containing ferromagnetic material were present in the oils. Taking advantage of this, a radiologically-safe, simple and inexpensive magnetic technique was applied to accelerate the natural precipitation of contaminants so that a cleaning effect was obtained. The technique is based on the use of small, inexpensive, permanent magnets. In this preliminary work a low energy configuration of magnets was used and even so a drastic reduction of specific activity of the oils and a high activity concentration in the sludge were obtained. Therefore a high-energy configuration will be used in the near future by means of which it is expected to increase the cleaning efficiency to be able to treat larger amounts of contaminated oils.

References

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