SEPARATION OF WASTE POLYETHYLENES BY DENSITY FRACTIONATION

J. Alrefai[1]

1: Petroleum & Petrochemicals Research Institute, KACST,

PO. Box 6086, Riyadh 11442, Saudi Arabia.
e-mail: alrefaie@kacst.edu.sa

ABSTRACT

A process is proposed for the separation of a mixture of polyethylene pellets scrap with different densities, contaminated with other impurities using a mixture of two or more miscible liquid components. The process is carried out at low pressure where one of these components is either a very volatile liquid (n-pentane) or a liquefied gas (n-butane, domestic gas) while the other component is a high density liquid such as glacial acetic acid or tetralin (1,2,3,4-tetrahydronaphthalene). The liquid mixture density could be decreased by increasing the concentration of the volatile component or gas pressure, or can be increased by aeration of the volatile liquid or lowering gas pressure. By this way all the fractions of the polyethylene mixture can be separated according to their densities.

Keywords: *Polyethylene pellets scrap, Separation by density, liquefied gas, n-pentane, acetic acid, Tetralin.*

الملخص

اقترحت في هذا البحث طريقة لفصل خليط من حبيبات البولي إيثلين المتخلفة صناعياً ذات الكثافات المختلفة والملوثة بالشوائب وذلك باستخدام مكونين سائلين أو أكثر قابلين للامتزاج . ثم إجراء العملية تحت الضغط العادي وباستخدام سائل فصل مكون من سائل طَيار مثل الـ n- بنتان أوغاز مُسَيل (n- بوتان ، غاز المنازل) وسائل مرتفع الكثافة مثل حمض الخل الثلجي أو التترالين (١ ، ٢ ، ٣ ، ٤ – رباعي هيدرو النفثالين) . يمكن خفض كثافة المزيج السائل بزيادة تركيز المٌكون الطيار أو ضغط الغاز ، أو رفع كثافته بتبخير السائل الطيار أوخفض ضغط الغاز . وبذلك يمكن فصل حبيبات البولي إيثلين الموجودة في المزيج السائل حسب كثافاتها .

1. INTRODUCTION

The amount of recycled plastics in 1993 was about 2% of all plastics produced in USA [Reisch, Marc S., 1995], and it is expected to grow to just over 3% by 1998. Of all the plastic resins, PET is number one in terms of post consumer recycling. However HDPE resin produced is four times more than PET, and in packaging HDPE resin is used more than twice as often [Glenn J., 1991]. The reason for this is that HDPE recycling industry has no regular supply of clean HDPE as much as PET. On the other hand in 1989, LDPE made up more than one-sixth of all plastic resin sold by USA firms and over half of that went to film markets. Recycled HDPE is usually used in making bottles, pipes, plastic bags and sheets used in households while recycled LDPE is usually used to make grocery bags. Limits to recycling plastics include the high costs and difficulty of collecting and sorting recyclable plastics according to their different materials. It is therefore envisaged that automated sorting techniques with an assist from a sensor system of some sort will ultimately help speed recycling. Therefore, one of the aims of this paper is to contribute to efforts directed at improving sorting techniques in plastic recycling.

A large quantity of polyethylene (PE) pellets scrap is produced, as a raw material with specification outside standard density grades, by the petrochemical industry around the world every year. This scrap contains different types of polyethylene pellets with varying densities that are difficult to sort into fractions with narrow density range. They are also contaminated with many types of impurities mixed with the pellets. This scrap is directed to be molded by plastic factories resulting in poor mechanical properties of the final products.

The polyethylene pellets scrap could be contaminated with the following [Perry, R. H., and Green, D. W., eds., 1984, and Super, M. S., et al., 1991]

Components	Range of density
High density PE:	0.940 - 0.965
Low density PE:	0.910 - 0.930
Glass:	2.400 - 2.800
Wood:	0.400 - 0.800
Sand, Gravel, dry, loose	1.400 - 1.700

The other aim of this paper is to propose a technique that can be used to separate this polyethylene (PE) pellets scrap from contaminants and from each other into different polyethylene fractions according to their various densities. This will improve the mechanical properties of the final products, and avoid the deterioration of processing equipment, leading to pure products similar to standard grades.

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[Super, M.S. et al., 1991 and 1992] proposed a process for the separation of waste thermoplastics from contaminants, and individual polymers from each other. This process is based on immersing the polymers in carbon dioxide and sulfur hexafluoride fluids and imparting high pressure near to their critical points. Separating the polymer fractions according to their densities was then carried by changing the pressure (i.e. the mixture density) of the fluids. However, this process suffers from working at high pressure (7.382 MPa with CO_2 and 3.759 MPa with SF_6) making this method very expensive for industrial application.

[Kenneth E.Kolb and Doris K. Kolb, 1991] suggested the use of liquids having different densities as a means for separating and identifying plastic materials. The liquids were used to separate two fractions, high density polyethylene from low density by flotation and sinking points of the plastic samples. They used for the separation of Low Density Polyethylene (LDPE) a mixture of 4:1 EtOH/H_2O, and for the separation of High Density Polyethylene (HDPE) a mixture of 1:1 EtOH/H_2O or 3:1 PrOH/H_2O. This method can't be useful for separating the PE with different close densities, and the recycling of liquid mixture components is not economical. In contrast to this, the separation process investigated in this work is carried out at or near atmospheric pressure and therefore is considered to be practical and economical to the industry.

2. MATERIALS AND METHODS

2.1. Materials

Four mixtures were used in these experiments: acetic acid and n-pentane; acetic acid with n-pentane and liquefied butane gas; acetic acid with n-pentane and liquefied domestic gas; and 1,2,3,4-tetrahydronaphthalene (tetralin) with n-pentane. The acetic acid was supplied by Fisher with 99.7% purity with a density =1.054 g/ml. The n-pentane used was 98% purity supplied by Winlab with a density = 0.626 g/ml. The liquefied n-butane was obtained from Air Products Company with 99.9% purity with a liquid density = 0.580 g/ml at its boiling point. The liquefied domestic gas was obtained from a local market supplier with a specific gravity of liquid (60/60 F^o) = 0.553 in summer, 0.535 in winter and 0.546 in other seasons. Tetralin was supplied by Fluka with purity >97% and a density = 0.973 g/ml. The PE pellets were standard grade polymers supplied by SABIC, Saudi Arabia. The pellets had a range of different densities as shown below:

PE1 (d.=0.918), PE2 (d.=0.921), PE3 (d.=0.924)
PE4 (d.=0.952), PE5 (d.=0.954), PE6 (d.=0.961)
PE7 (d.=0.965)

Each one of these polymer grades had a different shape than the others and therefore was visually identifiable.

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2.2. Method

Different standard grades of PE pellets were mixed and suspended in a liquid mixture with a predetermined density. The differences in densities between polyethylene pellets were used to separate them from each other using flotation and sinking principle by varying the density of the liquid mixture. The liquid mixture used for the separation is composed of two liquids as follows:

i) A liquid with a density more than HDPE (i.e. density > 0.965) such as acetic acid or tetralin.

ii) A low density liquid less than LDPE (i.e. density < 0.91) such as n-pentane or a liquefied gas such as n-butane or domestic gas.

2.2.1. Separation under liquefied gas pressure

A rig for separation experiments was constructed from PVC with a pipe of 1.5" diameter, and four ball valves. One valve is used as inlet for the mixtures and the others were used for collecting the liquid and PE mixtures from the pipe, as shown in (Figure 1) The PVC pipe was graduated and transparent to permit visual inspection of the separation process of PE pellets. The liquefied gas was injected from its cylinder through a 1/8" stainless steel tube into the PVC housing and was controlled by needle valves. A pressure sensor and a thermocouple with digital meters were installed through the PVC pipe. A leak valve for venting the liquefied gas was fitted to the top of the PVC pipe.

Two types of liquefied gases were used with this system: the n-butane gas and the domestic gas. Initially a liquid mixture of a high density liquid component with n-pentane was prepared so that its density was just above the HDPE. A known mixture of PE pellets containing different density pellets was placed with the initial liquid mixture through the inlet of the PVC pipe. The PE pellets floated completely on the liquid surface. The liquid density of the mixture was then lowered step by step by allowing small additions of the liquefied gas into the mixture and taking readings of the pressures for each liquid level (i.e. added volume) and observing the separated fractions of PE pellets as they sink. The sunk fractions were collected from the bottom of the pipe for verification of their shapes and densities. After separation of all PE fractions was achieved, the leak valve was opened to vent the gas from the mixture. It should be noted that in practical application a compressor could be used to re-condense the gases back in the gas cylinder.

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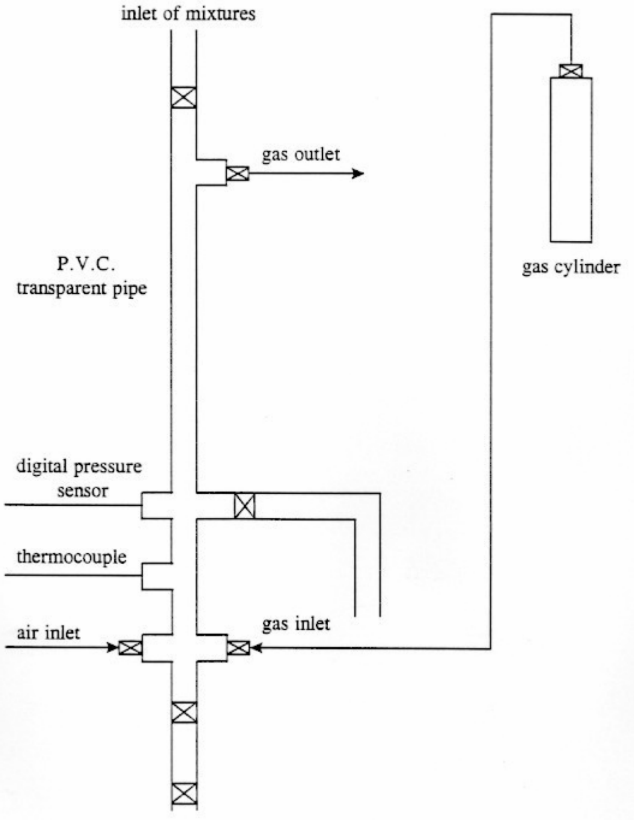

Figure 1 : experimental rig

2.2.2. Separation under atmospheric pressure

Two mixtures were used, acetic acid with n-pentane and tetralin with n-pentane. In the first mixture, experiments were conducted in the apparatus discussed above. An air injection inlet was added at the bottom of the apparatus to assist in the evaporation of the volatile liquid. With the second mixture, tetralin was found to be incompatible with the PVC, therefore a 50 ml graduated tube was used to conduct the separation experiments. The tube was fitted with a nozzle at the bottom for bubbling air through the solution. The relation between the density and the ratio of n-pentane with each mixture was determined first by weight. The separation of PE pellets fractions was achieved by first adjusting the liquid mixture density to make all the pellets float then the liquid density was lowered by sequential addition of the low density liquid component (i.e. n-pentane). The different fractions of PE pellets were separated as they sank. The mixture was well mixed by means of a magnetic stirrer. Following this separation, another PE pellets mixture was added to the liquid mixture. The PE pellets in this case sank and separated as they floated by increasing the liquid density using air bubbling in the liquid mixture to evaporate the light component. In real applications, the light component can be compressed and recycled.

3. RESULTS AND DISCUSSIONS

3.1. Separation Of PE Pellets Using A Mixture Of Acetic Acid / N-Pentane / Liquefied N-Butane Gas Mixture

The change of density with the volume % of n-pentane in acetic acid/n-pentane mixture was first determined by weight measurements, (Figure 2) and was found to fit an equation of $Y = -233.61\ X + 246.23$ where X is the mixture density and Y is the volume % of n-pentane. A mixture with a density of 0.97 was then prepared and placed in the experimental rig. This value was enough to float all the PE pellets.

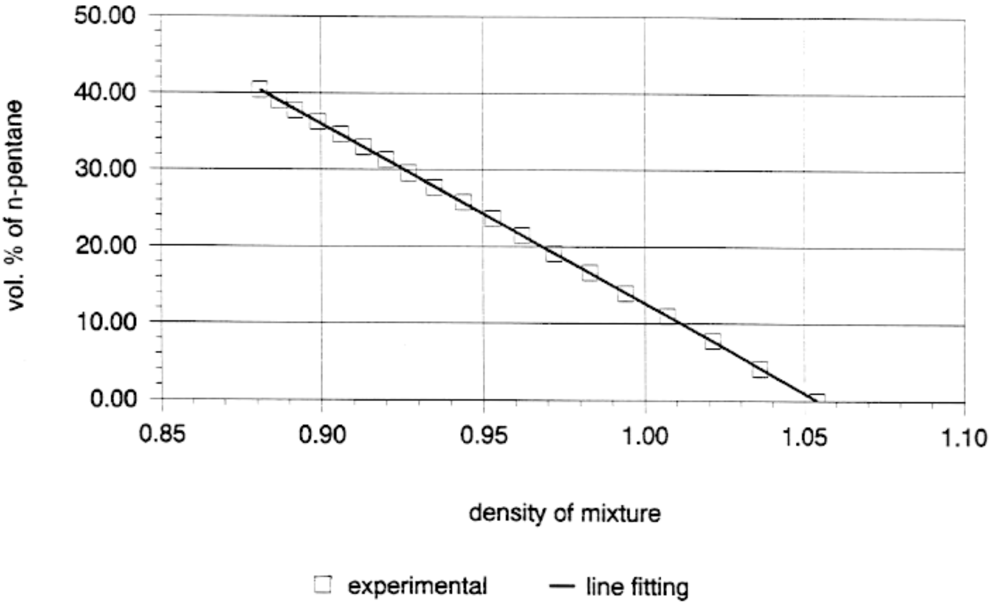

Figure 2: change of density with vol.% of n-pentane in acetic acid/n-pentane mixture

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The liquefied n-butane gas was then introduced into the mixture slowly. (Figure 3) shows that the added volume % of n-butane in the mixture increases linearly with pressure in the pipe as shown by the equation:

$Y = 1.2687\ P - 0.7625$

Where Y is the vol.% of liquefied n- butane in the mixture and P is the pressure.

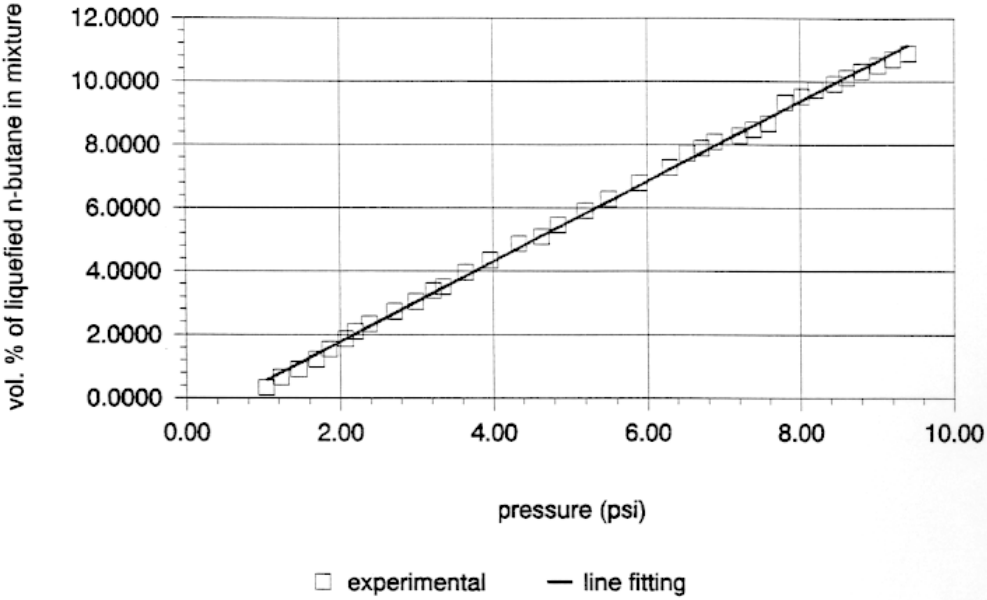

Figure 3: relation between gas pressure and volume % of liquefied n-butane in its mixture with acetic acid and n-pentane

This indicates that the density change of the liquid mixture is linear with pressure and volume % of the liquefied gas. This is shown in (Figure 4) where four fractions of PE pellets (PE1,3,4,7) were separated. Each fraction was collected from the bottom of the pipe for verification of the polymer density by color and shape of each fraction. With an n-pentane initial volume of 19.63 % in acetic acid /n-pentane mixture, the linearity between the density with pressure or the vol.% of liquefied butane gas was found to fit the equations,

$X = - 163.8691\ P + 161.5285$

Where X is the density and P is the pressure at separation or

$Y = - 189.9148\ X + 186.8356$

Where Y is the vol.% of liquefied butane and X is the density of PE pellets at separation.

It is noted that the pressures used for separation are much lower in comparison with pressures used by other workers [Super, M., et al. 1991].

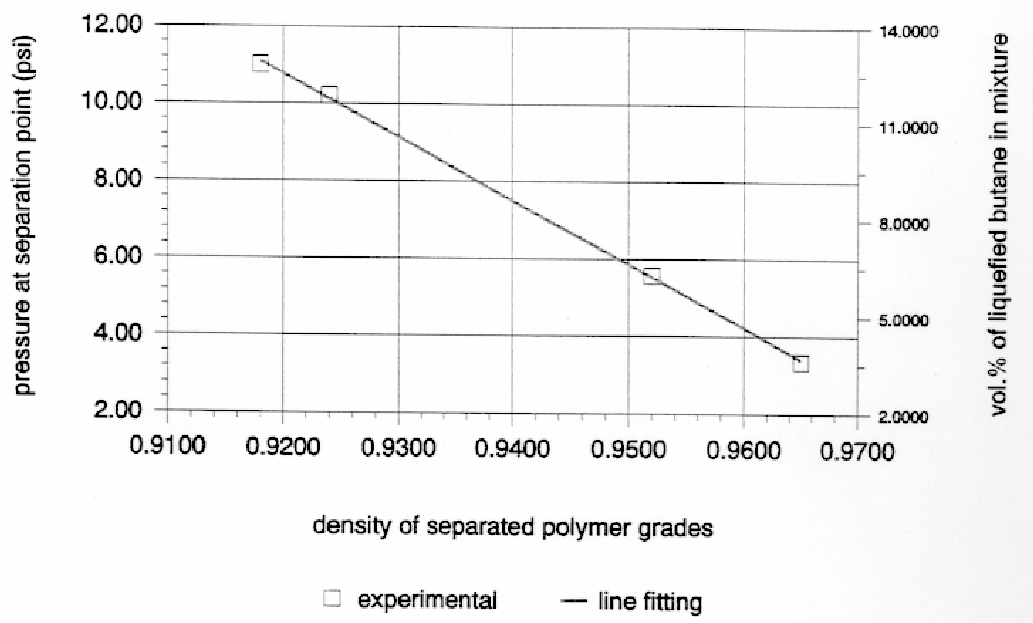

Figure 4: separation points of polyethylene pellets during addition of liquefied n-butane gas to acetic acid/n-pentane mixture

3.2. Separation of PE Pellets Using An Acetic Acid /N-Pentane /Domestic Gas Mixture

For economical reasons, domestic gas was used as a light component, easy to separate from the liquid mixture in this process. An initial liquid mixture containing acetic acid and n-pentane with a density of 0.990 was placed in the apparatus. The liquefied domestic gas was injected into the mixture. The relation between the injected volume % and pressure during injection is shown in (figure 5), which was found to fit the equation of $P = 1.6782\ Y - 1.3191$ where P is the pressure and Y is the volume % of liquefied domestic gas.

The separation of four different densities PE (PE1,3,4,7) according to the increasing pressure during the mixing of domestic gas with the liquid mixture was carried out. Each separated polymer fraction was collected at the bottom of the apparatus and removed for verification of its density by color and shape. A linear relation between the density of separated PE pellets with pressure or vol.% of liquefied domestic gas was obtained (figure 6).

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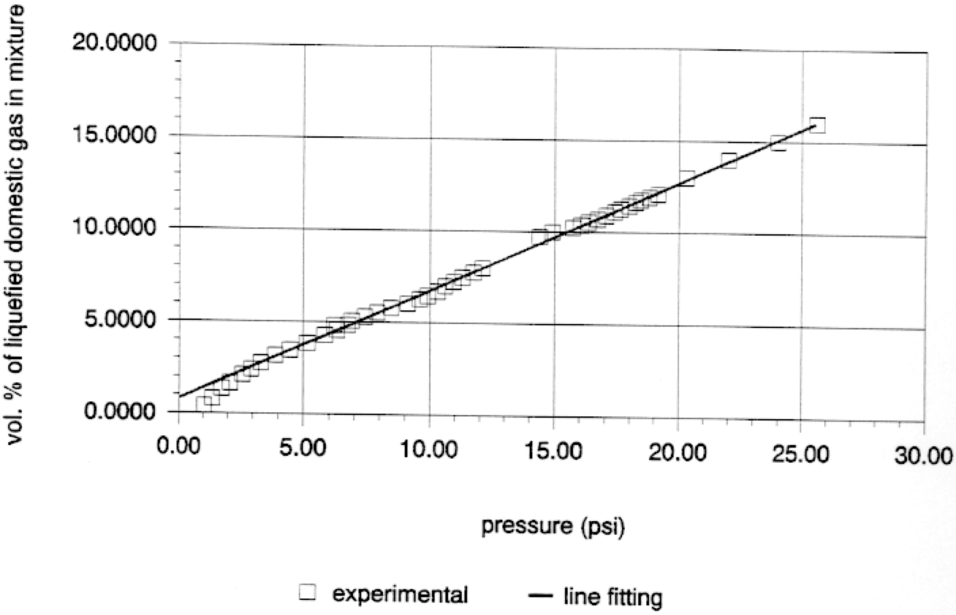

Figure 5: relation between gas pressure and vol.% of liquefied domestic gas in acetic acid/n-pentane mixture

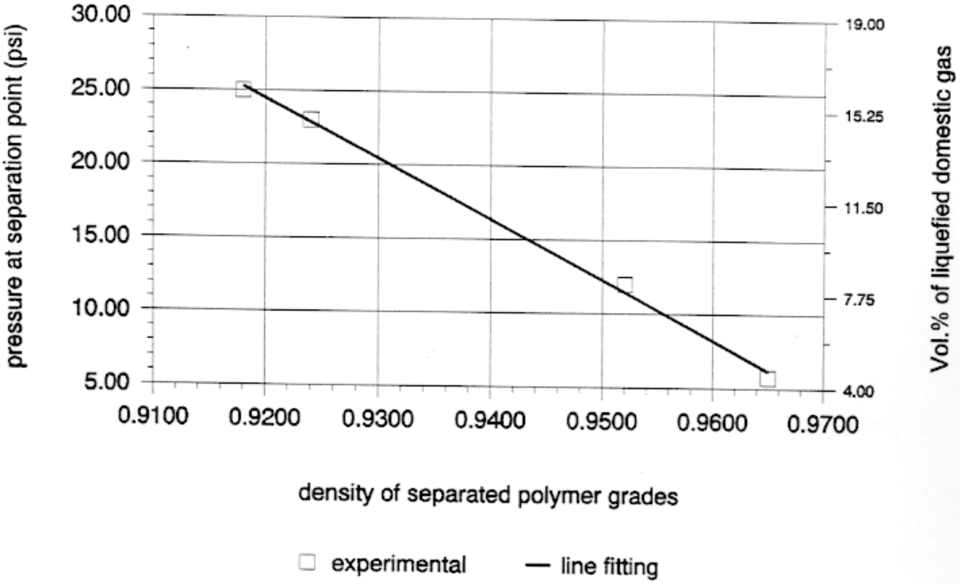

Figure 6: Separation points of polyethylene pellets during addition of liquefied domestic gas to acetic acid/n-pentane mixture

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A line was fitted to the experimental values with the equations

$P = -406.8351\ X + 398.7608$

where P is the pressure and X is the density of the separated PE. and

$Y = -\ 254.6829\ X + 250.4880$

Where Y is the vol.% of liquefied domestic gas and X is the density of separated PE. The reading of pressure values was taken directly after the introduction of liquefied gas.

The use of a small fraction of n-pentane with the acetic acid aims to prevent the change of glacial acetic acid to freeze when the temperature is low (its melting point is 16.2°C), and to prevent the corrosion of PVC apparatus.

3.3. Separation of PE Pellets Using a Mixture of N-Pentane/Acetic Acid

In the first step, the change of density of acetic acid and n-pentane mixture versus the volume percentage of n-pentane in the mixture was determined by weight (figure 2). A liquid mixture was prepared with a density suitable for floating six PE fractions (PE1-6). The liquid mixture density was lowered gradually by sequential addition of n-pentane and the PE pellets of each density was separated as shown in (figure 7).

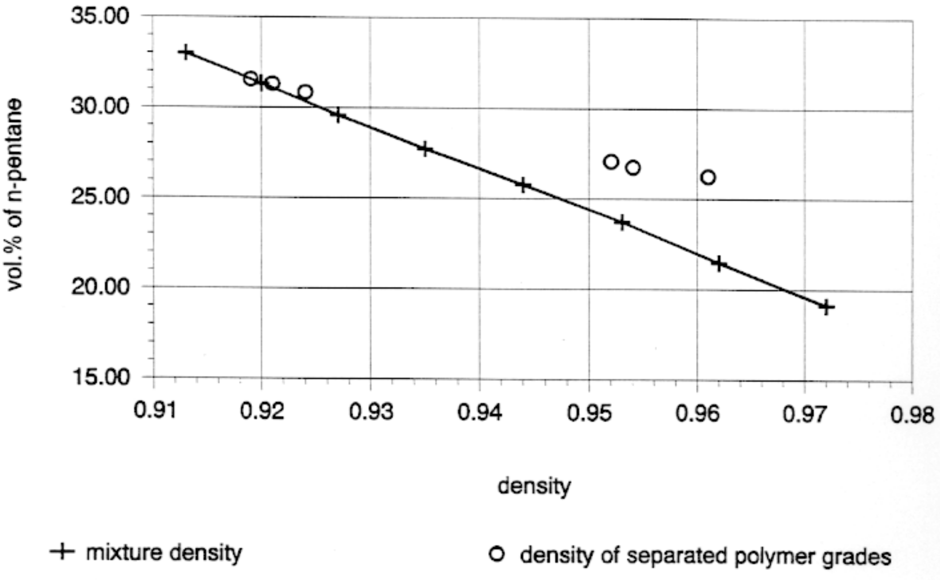

Figure 7: separation of polymer pellets with vol.% n-pentane in its mixture with acetic acid

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By bubbling air in the liquid mixture, it was possible to study the evaporation time of n-pentane from the mixture for three different air flow rates (figure 8). By this way, a new quantity of PE scrap can be separated by floatation.

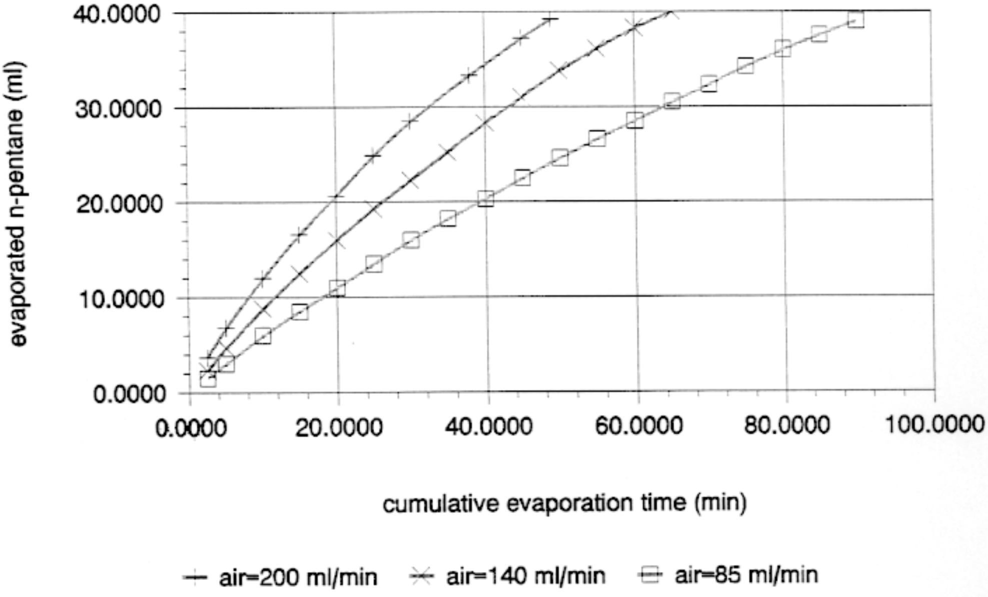

Figure 8: evaporation rate of n-pentane from a mixture of 40% V/V with acetic acid by air bubbling

3.4. Separation Of PE Pellets Using A Mixture Of N-Pentane/Tetralin

1,2,3,4-tetrahydronaphthalene was used instead of acetic acid due to its suitable density. Since tetralin is corrosive to PVC, the experiment was conducted in a graduated glass tube. A liquid mixture with a density suitable for floating six PE fractions (PE1-6) were prepared. The relation between the density with vol.% of n-pentane in its mixture with tetralin fitted the line equation

$$Y = - 293.49 X + 283.80$$

where Y is the volume % of n-pentane and X is the density of mixture (Figure 9).

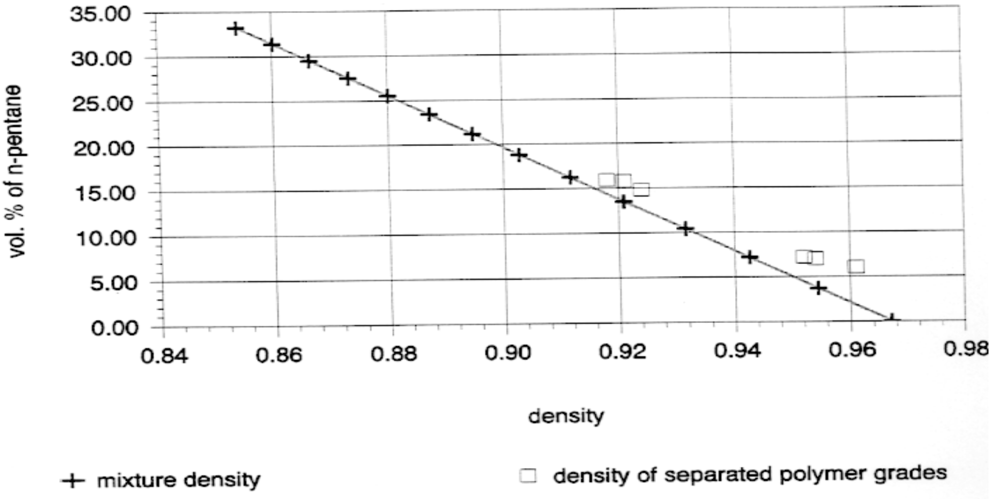

Figure 9: separation of polymer pellets with vol.% n-pentane in its mixture with tetralin

4. CONCLUSION

The results presented in this paper show that by changing a liquid density in which different PE fractions are placed, it was possible to separate these pellets into their original fractions of different densities. Lowering or increasing the density of the liquid mixtures was done by adding or evaporating a volatile liquid or liquefied gas from the liquid mixture. n-Pentane was used as the volatile liquid with acetic acid or with tetralin. Liquefied n-butane and domestic gas were used with acetic acid/n-pentane mixture and the liquid mixture density was varied by changing the gas pressure up to 25 psi. The liquids used in the separation are cheep and recyclable, and the conditions of the separation were at or near atmospheric pressure.

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