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A Study on Recycling of Waste Polyethylene Film

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폐폴리에틸렌 필름의 재활용에 관한 연구

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Abstract The compounds of recycled polyethylene(PE) and fly-ashes were prepared. Polymers used were sorted PE from mixed plastics of household waste and Low Density Polyethylene(LDPE) and Linear Low Density Polyethylene(LLDPE) recycled from the scrap of packaging film plants. Fly-ashes were from the power plant and from the household waste incinerator. The tensile strength of recycled LDPE and LLDPE compounds decreased and the flexural modulus increased with greater amount of the power plant fly-ash. Anthracite fly-ash gave rise to slightly higher tensile and flexural strength of the LLDPE mixtures than bituminous coal fly-ash presumably due to higher content of unburned carbon. The incinerator fly-ash introduced to household waste PE enhanced both tensile strength and flexural modulus of the compounds. When LDPE and household waste PE were used together, the synergistic effect of incinerator fly-ash to household waste PE was offset by reduced crystallization of LDPE due to the filler particle. The compounds of household waste PE and incinerator fly-ash might be applied to structural materials for such as sewage pipe, which reduces the waste treatment cost and conserve the environment and resources.

Key Words : Recycling, Waste polyethylene, Compounds, Tensile strength, Flexural modulus

요 약 폐폴리에틸렌 필름을 재활용하기 위하여 폐폴리에틸렌과 비산재를 사용하여 콤파운드를 제조하였다. 실험에 사용한 고분자는 생활쓰레기에서 분리수거한 폴리에틸렌(PE)과 포장용 필름을 생산하는 공장에서 발생하는 스크랩에서 얻어지는 저밀도폴리에틸렌(LDPE)과 선형저밀도폴리에틸렌(LLDPE)이다. 폐폴리에틸렌의 물리적 성질을 개량시키기 위하여 화력발전소 굴뚝에서 포집한 석탄재와 생활쓰레기 소각로굴뚝에서 얻어지는 비산재를 사용하였다. 발전소 석탄재를 투입하면 재활용 LDPE와 LLDPE 콤파운드의 인장강도는 감소하고 굴곡모듈러스는 증가하였다. 역청탄재를 혼합한 경우에 비하여 무연탄재를 투입한 LLDPE 콤파운드의 인장강도와 굴곡강도가 약간 높았으며, 이는 무연탄재가 다소 많은 량의 미연소 탄소를 가지기 때문으로 사료된다. 생활쓰레기에서 얻어진 PE 단독에 소각로 비산재를 투입하면 인장강도와 굴곡강도가 모두 증가하였다. 이와 같은 소각로 비산재에 의한 상승효과는 생활쓰레기 PE에 포장용 필름 스크랩의 LDPE를 섞게 되면 Filler 입자가 LDPE의 결정화를 방해하기 때문에 상쇄되는 현상이 관찰되었다. 소각로 비산재를 혼합하여 생활쓰레기 PE를 재활용한 콤파운드는 하수관 등을 제조하는 원료로 사용이 가능하다. 본 연구결과를 폐플라스틱의 재활용에 적용시키면 폐기물처리 비용을 절감하고 환경보존과 자원절약에 기여할 수 있다.

1. Introduction

Plastics are essential in the industrialized society and their usage increased tremendously in various fields in past decades, which reflects economical production and superior properties of polymers. Polymeric materials are widely used as consumables of household appliances and industrial resources of packaging, construction, transportation, automobiles, electronics and agricultural area. Therefore huge amount of waste plastics is inevitable and environmental problems arise since plastics are not easily degradable.

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Table 1. Output and recycling of waste plastics for commodity resin in Korea (Unit: Metric Thousand Tons).

Year	Amount of Synthetic Resin				Total Output of Waste Plastics	Output of Household Waste Plastics	Amount of Recycled Waste Plastics	Percentage of Recycling
	Production	Import	Export	Domestic Demand				
1992	5,169	166	1,957	3,378	1,943	1,154	173	9 %
1997	7,261	121	3,696	4,046	2,800	1,400	500	18 %
2002	9,534	189	4,875	4,848	3,650	1,500	908	25 %

Table 2. Domestic demand of synthetic resin in Korea in 2003.

	LDPE	HDPE	PP	PS	ABS	PVC	Total
Demand (Metric Thousand Tons)	867	811	1,261	503	236	936	4,614
Occupancy Rate (%)	18.8	17.6	27.3	10.9	5.1	20.3	100

Table 1 shows the output and recycling of waste plastics for commodity synthetic resin in Korea [1]. The domestic consumption of plastics recently amounts to 5 millions tons and the output of waste plastics is about 3.7millions tons but the portion of recycling remains merely 25%, which suggests that the disposal of waste plastics is dominated by landfill and incineration, resulting in serious environmental problems and waste of useful resources. Table 2 shows the demand of commodity resin in Korea in 2003 [2]. Polyolefin including polyethylene(PE) and polypropylene(PP) occupies 64% and PE does 36%. Therefore recycling of PE is very important above all.

The collected plastics of post-consumer are usually mixtures of different types of polymers and may contain plasticizer, antioxidant and a variety of additives depending on the applications. The heterogeneity of commingled plastic frequently tends to phase separation on processing, and deteriorates physical and mechanical properties of the recycled products [3, 4]. In order to mitigate the deterioration, one may add quinone methide, phosphate, sulfuric oxide, hindered amine stabilizer(HAS) to recycled plastics [5-8]. A compatibilizer may suppress phase separation; ethylene- propylene random copolymer (EPR), poly(styrene-block-(ethylene-co-butylene-1)-block styrene) triblock copolymer (SEBS), styrene-butadiene-styrene (SBS) triblock copolymer, polystyrene(PS)-block-polybutadiene are recommended for recycled polyolefin [9-11]. A reactive compatibilizer would be more aggressive in reducing phase separation because it creates covalent bonds between different polymers [12, 13]. However it is too much expensive for recycling of commingled plastic of post-consumer. Thermoplastic resin is recyclable and inexpensive compared with metals or engineering plastics, but it is weak and thermally instable. The shortcoming may be circumvented by adding filler, plasticizer, toughening agent and flame-retardant. Fillers are rarely used with PE since they interfere with the crystallinity of the polymer to give rather brittle products of low ductility. Carbon black has some reinforcing effect and is useful in cross-linked PE [14]. Somewhat better results may be achieved by using surface treated fillers with silane and titanate coupling agents and compounds of increased rigidity and tensile strength compared with unfilled polymers may be obtained.

The objectives of this paper are recycling waste PE and enhancing rigidity and tensile strength of compounds by introducing fillers to recycled PE. PE used was the sorted plastic from household waste and the relatively clean scrap derived from packaging film plants. Fillers were fly-ashes from the power plant and from the incinerator of household waste. The compounds obtained in the study might be applied to structural materials for such as sewage pipe, rainwater duct, and irrigation for agriculture.

2. Experimental

2.1 Materials

Recycled PE and fly-ashes from the power plant and from the incinerator of household waste were used. The source of PE was the scrap derived from packaging film plants and the sorted plastic from household waste. The collected polymer lump was masticated intensively twice by a twin-screw extruder and pelletized before compounding. The PE film scrap obtained from film plants was relatively clean and not contaminated. And two different types of recycled PE containing about 60~70 % of Low Density Polyethylene(LDPE) and Linear Low Density Polyethylene(LLDPE) with remaining components of mostly Nylon 6, Poly(ethylene terephthalate)(PET), PP were used respectively. Recycled PE obtained from household waste was somewhat heterogeneous and mixed with aluminum foil, paper, and other contaminants.

Table 3. Composition of fly-ash obtained from power plant in percentage after complete burning.

Component	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	SO_3	TiO_2	Others
Anthracite Fly-Ash	54.60	31.78	4.58	0.87	0.78	0.23	4.03	0.26	1.91	0.96
Bituminous Coal Fly-Ash	55.23	21.26	8.54	5.50	1.29	0.48	1.09	3.63	1.37	1.61

Table 4. Composition of fly-ash obtained from incinerator of household waste.

Component	SiO_2	CaO	Al_2O_3	Fe_2O_3	Na_2O	K_2O	ZnO	MgO	Others
Weight %	26.93	17.86	11.30	1.72	3.67	2.92	1.57	8.79	25.24

Fillers were fly-ashes from the power plant and from the incinerator of household waste. The fly-ashes of anthracite and bituminous coal were supplied from the power plant. The average particle size of the power plant fly-ash was in the range of 20~30 mm and the content of unburned carbon was about 14~20 % for anthracite and about 3~4 % for bituminous coal. The composition of the power plant fly-ashes after complete burning in an electronic furnace is shown in Table 3. The particle size of the incinerator fly-ash was similar to those of the power plant. The representative composition of the incinerator fly-ash is shown in Table 4, in which others may consist of heavy metal compounds and varying amount of soot.

2.2 Compounding and Preparation of Test Specimens

Recycled PE pellet and fly-ash powder were weighed to obtain a definite composition. They were dry mixed at the atmosphere and compounded with a twin-screw extruder(Brabender Compounder DSK 42/7) to make a rodlike strand, and cut again to give small pellets with a pulverizer. The extruder was operated at the screw speed of 60 rotation per minute and with the temperature profile of 170~172℃ for zone 1(near hopper), 200~201℃ for zone 2, and 214~216℃ for zone 3, varied with different composition of compounds. The amounts of fillers introduced were 0, 5, 10, 15 and 20 % for a series of compounds.

Test specimens were prepared with an injection molder(Newbury HV1-25ARS). The specimens for flexural and tensile tests were fabricated with the dimension shown in Fig. 1. The cooling water temperature was 50℃ and the mold time was maintained at 1 to 2 minutes during injection molding. The temperature profile of the molder was varied to obtain an optimum specimen. The typical temperatures were 282~286℃ for rear zone, 269~271℃ for front zone, and 255~267℃ for nozzle.

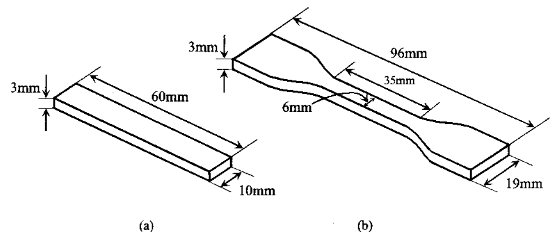

Fig 1. Dimension of injection-molded specimens for flexural test(a) and for tensile test(b).

2.3 Measurement of Mechanical Properties

Tensile properties were tested with an Instron 4466(Series IX, V1022) at the crosshead speed of 50mm/min and at 5~7℃ following the method of ASTM D 638. Tensile tests were repeated 7 times for each specimen and averaged with 5 measurements to obtain tensile strength and modulus discarding a maximum and a minimum. Flexural tests were performed by an Instron 6022 following the method of ASTM D 790. All specimens were stored at 23±2℃ and relative humidity of 50±5 % for more than 40 hours before measurement. Flexural tests were repeated 5 times for each specimen and averaged out to obtain flexural modulus and strength. The variation of the individual measurements for each specimen was within about 5% for tensile tests and about 3% for flexural tests.

3. Results and Discussion

The tensile strength, tensile modulus, flexural modulus, and flexural strength for the compounds of recycled PE from the film scrap and the power plant fly-ash are shown in Fig. 2. All mechanical properties for LDPE composite are superior to those for LLDPE composite, which is attributed to crystallinity of polymer. Because LLDPE contains the comonomer of butane, hexane, or octane, the LLDPE chain has less regular structure than the LDPE chain resulting in lower crystallinity. As the power plant fly-ash contents increases, the tensile strength of compounds decreases in Fig. 2(a) and the tensile modulus increases in Fig. 2(b), which suggests the dual effects of the fly-ash particles. Thus fly-ashes interfere with crystalllizability of PE reducing tensile strength and reinforce the polymer compounds increasing tensile modulus [14]. The effect of fly-ashes on the tensile and flexural properties of the compounds is larger in LDPE than in LLDPE, which is again related to higher crystalllizability of LDPE. The difference between the effects of anthracite fly-ash and bituminous coal fly-ash is minor. The tensile and flexural strengths of LLDPE compounds containing anthracite fly-ash are slightly higher than the corresponding of bituminous coal fly-ash as can be seen in Fig. 2(a) and Fig. 2(d). This would be resulting from higher contents of unburned carbon in anthracite fly-ash.

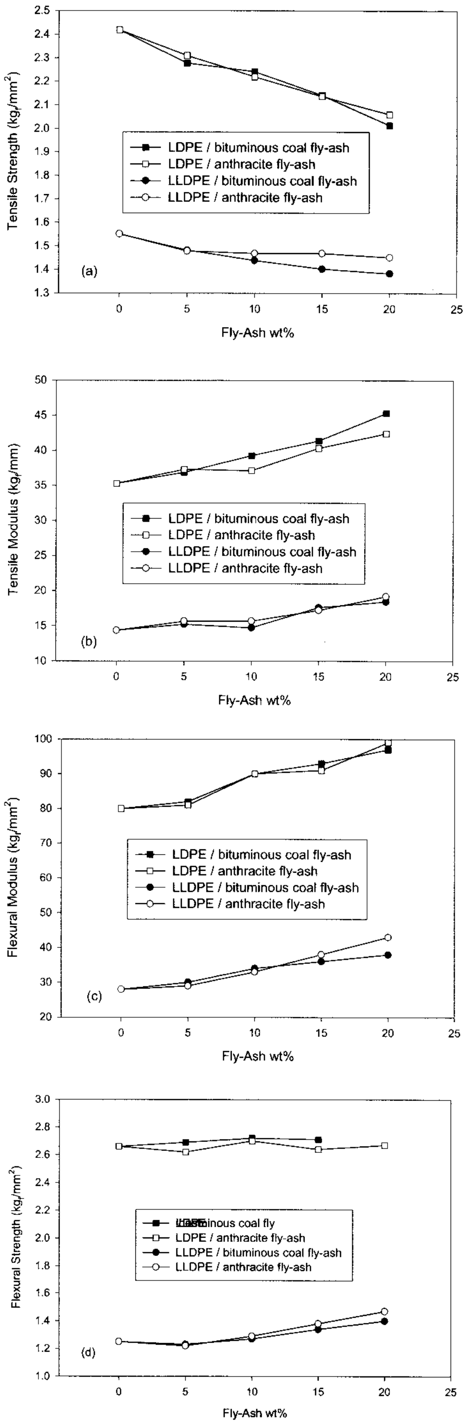

Fig 2. Tensile strength(a), tensile modulus(b), flexural modulus(c), and flexural strength(d) for compounds of recycled PE from film scrap and power plant fly-ash as a function of filler content.

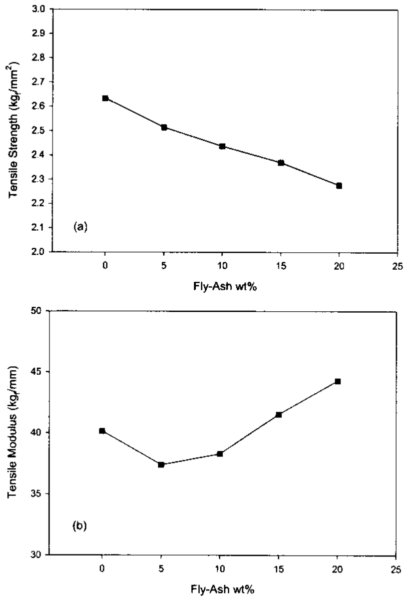

Fig 3. Tensile strength(a) and tensile modulus(b) for compounds of recycled LDPE from film scrap and incinerator fly-ash as a function of filler content.

Table 5. Tensile strength, tensile modulus, flexural modulus, and flexural strength of the compounds recycled from waste polyethylene film.

Composition of Compounds		Tensile Strength (kg_f/mm^2)	Tensile Modulus (kg_f/mm)	Flexural Modulus (kg_f/mm^2)	Flexural Strength (kg_f/mm^2)
Composition of Recycled Resin	Filler Contents				
LDPE	0 %	2.63	40.15	84	2.74
	Incinerator Fly-Ash 5%	2.51	37.38	83	2.69
	Incinerator Fly-Ash 10%	2.44	38.28	83	2.65
	Incinerator Fly-Ash 15%	2.37	41.53	90	2.68
	Incinerator Fly-Ash 20%	2.28	44.29	105	2.85
Household Waste PE	0 %	1.17	15.20	27	1.13
	Anthracite Fly-Ash 10%	1.14	17.21	34	1.30
	Incinerator Fly-Ash 10%	1.23	19.39	37	1.36
	Incinerator Fly-Ash 20%	1.29	26.75	62	1.65
30/70 LDPE/ Household Waste PE	0 %	1.66	21.27	38	1.42
	Anthracite Fly-Ash 10%	1.55	23.54	44	1.48
	Incinerator Fly-Ash 10%	1.58	25.94	44	1.48
50/50 LDPE/ Household Waste PE	0 %	1.93	24.14	52	1.71
	Anthracite Fly-Ash 10%	1.79	28.81	55	1.75
	Incinerator Fly-Ash 10%	1.86	28.48	59	1.83
	Incinerator Fly-Ash 20%	1.83	32.85	70	1.99

The mechanical properties for the compounds of recycled LDPE and incinerator fly-ash are shown in Fig. 3 and in Table 5. The tensile strength behavior is similar to the cases of the compounds containing the power plant fly-ash as shown in Fig. 3(a). However, the tensile modulus of LDPE is decreased by introducing 5 % of incinerator fly-ash and tends to increase with higher contents of fly-ash in Fig. 3(b), which is quite different from those with the power plant fly-ash in Fig. 2(b). It is understood that the interaction between LDPE and a certain component in the incinerator fly-ash affected the miscibility of the compounds. In fact we were able to obtain a good specimen of LDPE compounds with 5% incinerator fly-ash by increasing temperatures and extending mold time on injection molding. When the interconnected bi-continuous structure is formed by spinodal decomposition or a favorable interaction between components exists, the viscosity of the multi-component system would be increased [4]. The tensile modulus for the compounds of recycled LDPE filled with 10 % or greater amount of incinerator fly-ash seemed to be influenced by reinforcing effect of filler particles in Fig. 3(b).

Properties of unfilled polymer blends of LDPE recycled from the film scrap and PE from the household waste plastics can be discussed in Table 5. Tensile and flexural properties increased always with higher composition of LDPE. It is interesting to note that the tensile strength of the household waste PE compounds is decreased by introducing anthracite fly-ash but is slightly increased by incinerator fly-ash compared with unfilled polymer. For comparison with different type of fly-ash the corresponding backgrounds were darkened in Table 5. The modulus of the polymer compounds is increased by both fillers and the effect is greater with incinerator fly-ash than with anthracite fly-ash. Thus incinerator fly-ash is more beneficial than anthracite fly-ash since it is capable of increasing both tensile strength and rigidity of PE recycled from the household waste plastics. The similar effects of filler are observed for 30/70 and 50/50 polymer blends of LDPE and household waste PE. The tensile strength and modulus of the compounds filled with 10 % fly-ash are higher with incinerator fly-ash than with anthracite fly-ash in Table 5. In contrast to the cases of household waste PE compounds without LDPE, as the composition of LDPE increased in the recycled polymer blends the tensile strength of the compounds is reduced by introducing fillers, which is related to crystallinity of PE discussed with compounds of LDPE and incinerator fly-ash.

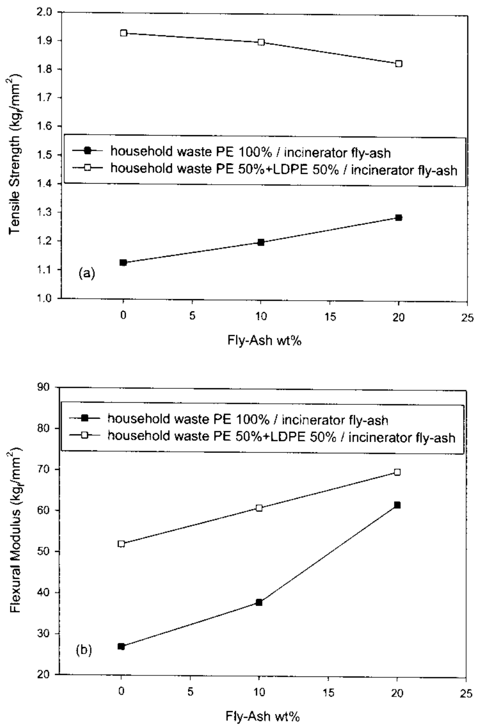

Fig 4. Tensile strength(a) and flexural modulus(b) for compounds of recycled PE and incinerator fly-ash as a function of filler content.

In summary performance enhancement by introducing incinerator fly-ash is most pronounced in the household waste PE. With greater amount of incinerator fly-ash both tensile strength and flexural modulus of the compounds are increased in the household waste PE but lower tensile strength and higher flexural modulus are obtained in 50/50 recycled polymer blends of LDPE and household waste PE in Fig. 4. The household waste PE may consist of mixed plastics, hard to recycle, and is usually incinerated or land-filled. The incinerator fly-ash is very harmful and should be disposed properly. Thus the compounds of the waste PE and incinerate fly-ash is the combination of wastes. They might be applied to structural materials for such as sewage pipe, which reduces the waste treatment cost and conserve the environment and resources. When the compounds properties are not enough to apply to a specific use, it would be desirable to consider mixing virgin polymer or placing the recycled compounds in between multi-layered composites, for instance the compounds of waste PE and incinerate fly-ash inside with virgin polymer outside. The tensile strength and the rigidity of sewage pipe are required to be above 200kg$_f$/㎠ and 3.5kg$_f$/㎠, respectively, by Korean Agency for Technology and Standards as specified in GR M3006. Finally it should be noted that the incinerate fly-ash is detrimental to health because it is fine powder and may comprise heavy metals and poisonous stuffs derived from household waste. The equipments for manufacturing the compounds should be modified to handle the incinerate fly-ash safely.

4. Conclusions

The tensile strength of recycled LDPE and LLDPE compounds decreased and the flexural modulus increased with greater amount of the power plant fly-ash. The tensile strength and rigidity of LDPE compounds were superior to those for LLDPE compounds, which was attributed to higher crystallinity of LDPE. Anthracite fly-ash gave rise to slightly higher tensile and flexural strength of the LLDPE mixtures than bituminous coal fly-ash presumably due to higher content of unburned carbon. The incinerator fly-ash introduced to household waste PE enhanced both tensile strength and flexural modulus of the compounds. When LDPE and household waste PE were used together, the synergistic effect of incinerator fly-ash to household waste PE was offset by reduced crystallization of LDPE due to the filler particle.

The compounds of household waste PE and incinerator fly-ash might be applied to structural materials for such as sewage pipe, which reduces the waste treatment cost and conserve the environment and resources.

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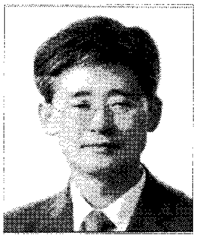

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<Research Interest>
Polymer Engineering, Recycling of Polymer