

Waste Tire Granular Modification Using Gamma Radiation as Concrete Aggregate

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ABSTRAK

Ban bekas kendaraan bermotor merupakan masalah yang serius dan rumit karena sulit terurai di lingkungan, tidak dapat dibentuk ulang (termoset), dan sulit didaur ulang. Tujuan penelitian ini adalah untuk mempelajari pengaruh modifikasi granul limbah ban menggunakan radiasi gamma sebagai agregat beton. Granul ban 4 mesh ($\leq 4,8$ mm) dikenai dosis radiasi gamma yang bervariasi (20, 40, dan 60 kGy). Pengaruh radiasi gamma terhadap granular ban diidentifikasi melalui perubahan gugus fungsi, densitas ikatan-silang, dan densitasnya. Beton berbentuk kubus (15 cm) dicetak dengan mencampur bahan baku seperti semen, pasir, kerikil, air, dan granular ban dalam berbagai kandungan. Beton kering (usia 6 hari) direndam dalam air selama 1 hari untuk menentukan daya serap airnya. Beton usia 7 hari diukur kuat tekannya menggunakan mesin uji tekan. Hasil penelitian menunjukkan pemaparan radiasi gamma pada 40 kGy dapat meningkatkan densitas ikatan-silang granular ban, yang menunjukkan bahwa ikatan-silang polimer berhasil dicapai. Beton K100 dengan granular ban teriradiasi 40 kGy memberikan peningkatan kuat tekan 150% (104 kg/cm^2) dibandingkan dengan granular ban non-iradiasi (69 kg/cm^2). Walaupun beton dengan granular ban teriradiasi memberikan kuat tekan yang lebih rendah dibandingkan dengan beton tanpa granular ban, beton dengan granular ban teriradiasi kadar optimum (4% b/b) masih memenuhi standar SNI 03-0349.

Kata kunci: Agregat beton, Granul ban bekas, Radiasi gamma

ABSTRACT

Automotive waste tires are a serious and complicated issue because it is difficult to degrade in the environment, cannot be reshaped (thermosets), and are difficult to recycle. The aim of this work is to study the effect of granular modification of waste tires using gamma radiation as a concrete aggregate. Tire granular 4 mesh (≤ 4.8 mm) is exposed by varied gamma radiation doses (20, 40, and 60 kGy). Gamma radiation effect on tire granular is identified through the functional groups, cross-link density, and density. Cubical concrete (15 cm) is molded by mixing raw materials such as cement, sand, gravel, water, and tire granular in various content. Dry concrete (6 days curing) is hydrated in water for 1 day to determine the water absorption. Concrete at 7 days curing is measured the compressive strength using compression testing machine. The result shows exposing gamma radiation at 40 kGy can increase the crosslink density of tire granular, which indicates polymer crosslinking is successfully achieved. Concrete K100 with irradiated tire granular at 40 kGy gives compressive strength improvement 150% (104 kg/cm^2) compared to non-irradiated tire granular (69 kg/cm^2). Even though concrete with irradiated tire granular gives lower compressive strength compared to concrete without tire granular, concrete with irradiated tire granular optimum content (4%) are still fulfill standard of SNI 03-0349.

Keyword: Concrete aggregate, Gamma radiation, Tire waste granular

1. INTRODUCTION

One of the important environmental issue is the end-of-life of automotive waste tires. However, there is a lack of information on tire final disposal management issues. Innovative solutions are invented to address the issue of tire disposal such as recovery program, demonstrating the recycling advantages, and life cycles assessments update. Likewise, it is important to consider how waste tires can be transformed into a worthwhile resource [1]. Waste tires recycling has been performed such as by common method is convert it to produce vapor, heat, or electricity. Waste tires are used for utilizing alternative fuel in cement kilns, for producing high carbon powder, and producing pyrolytic gas [2, 3, 4]. Unfortunately, these methods lead to the production of highly polluting byproducts such as hydrocarbon gas, volatile compounds, heavy and light oils, zinc oxide, and zinc sulfide [5]. Another approach is used waste tires as filler in floor mats and carpet padding. Recently, waste tires are used as substitute of fine or coarse aggregate in concrete [6, 7, 8]. Granulated tire waste has been mixed in concrete, which give acceptable workability, lower compressive strength and unit weight, higher air content, and constant concrete porosity [7]. High tire chips content (15%) in concrete gives diminution density and compressive strength [8].

While there are various advantages of adding tire granules to concrete to increase its toughness, there are also some drawbacks, such as lower compressive strength values, that should be considered. One approach is the use of gamma radiation [9]. As radiation from a gamma ray reacts with a polymer material, the polymer material absorbs the energy and generates active species such as radicals, thus causing different chemical reactions. The basic processes that are the outcomes of such reactions involve crosslinking, chain scission, oxidation, long-chain branching, and grafting. Polymer crosslink scheme principally consists of radical formation, initiation, propagation, and termination (Figure 1) [10].

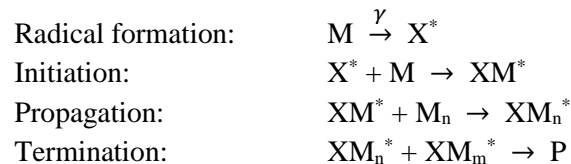


Figure 1. Polymer crosslink scheme
(M = polymer, * = radical, P = crosslinked polymer)

Few researchs are regarding to apply gamma irradiation to modify polymer crosslink properties [10], rubber crosslink [11], and the physical and mechanical polymer concrete properties [12, 13]. Modification of tire fibers using varied gamma radiation dose (50-300 kGy) has been studied [14, 15], which higher dose lead to reduce compressive strength of concrete. By using gamma radiation, recycled high-density polyethylene and ground tire rubber blends can be functionalized through higher interaction between elastomer and acrylamide functional groups; this allows improvement of their mechanical properties for doses from 25 to 50 kGy [16]. Another study show butyl rubber (the type of rubber in tires) undergoes chain scission when exposed to gamma radiation at a dose of 45 kGy [17].

Given the earlier results, the goal of this work is to modify tire granular using low gamma radiation (20-60 kGy) for improvement of concrete properties. Hopefully, this work is undoubtedly a valuable option for reducing environmental pollution without improper disposal.

2. METHODOLOGY

This work consisted of several processes, including tire granular preparation, tire granular irradiation, concrete molding, and tire and concrete granular testing. Table 1 provides a list of the materials used to compose concrete. Methanol and toluene are also used for tire granular properties testing before and after irradiation process. Several equipment is used in this study included analytical balance, cubical concrete mold (15 cm), gamma irradiator Co-60 Type 1, spectrometer fourier transform infrared spectroscopy (FTIR) Shimadzu IR Spirit, compression testing machine, pycnometer, oven, and other glassware.

Table 1. Concrete formulation

No.	Ingredients	Content (% w/w)				
		A	B	C	D	E
1	Cement	10	10	10	10	10
2	Gravel	41	41	41	41	41
3	Water	13	13	13	13	13
4	Sand	36	34	32	30	28
5	Tire Granular	0	2	4	6	8

2.1 Tire Granular Preparation, Irradiation, and Testing

Tire granular is obtained from the results of chopping waste tires. Granular tires were sieved using 4 mesh sieve (≤ 4.8 mm). The granular tires are inserted to an irradiator and exposed by varied gamma radiation doses (20, 40, and 60 kGy).

Functional groups of before and after irradiated tire granular were characterized using spectrometer FTIR at wavelength range 4000-700 cm^{-1} . Density of before and after irradiated tire granular were measured using pycnometer.

Crosslink density before and after irradiated tire granular were measured using previous method by Blume and Kiesewetter [18]. Sample (tire granular) 0.05 gram was hydrated in toluene solution for four days (until equilibrium) at room temperature (25°C). Tire granular was dried to a constant weight for four days using an oven (60°C). Sample crosslink density was calculated using Flory-French equation (Eq. 1). Rubber volume fraction in the solvent (V_r) in Eq. 1 is calculated using Ellis dan Welding (Eq. 2). In Eq. 1, ν represents crosslink density per volume (mol/cm^3), V_0 represents solvent molar volume (toluene: 106.9 cm^3/mol), and χ represents interaction parameter of Flory-Huggins (0.393). In Eq. 2, D represents the weight of the dried sample, \emptyset represents the weight fraction of the filler, W represents the initial weight of the sample, A_0 represents the quantity of absorbed solvent (toluene), ρ_r represents sample density, dan ρ_s represents solvent density (toluene: 0.867 g/mL).

$$\nu = - \frac{\ln(1-V_r)+V_r+\chi V_r^2}{V_0 \left(V_r^{\frac{1}{3}} - \frac{V_r}{2} \right)} \quad (1)$$

$$V_r = \frac{(D-\emptyset W)\rho_r^{-1}}{(D-\emptyset W)\rho_r^{-1}+A_0\rho_s^{-1}} \quad (2)$$

2.2 Concrete Molding and Testing

Cement, sand, gravel, tire granular, and water were weighed according to the formulations in Table 1. The materials were mixed and inserted in the cubical mold. The concrete was removed from the mold after 1 day curing (Figure 2). Concretes were dried (cured) at room temperature until 6 days curing.



Figure 2. Concrete at 1 day curing

The water absorption of concrete (w_a) was determined by measuring the concrete weight before (m_a) and after (m_b) hydrated in water for 24 hours. The water absorption of the concrete was calculated using Eq. 3. The compressive strength of the 7 days cured concrete was measured using a compression testing machine.

$$wa = \frac{(m_b - m_a)}{m_a} \times 100\% \quad (3)$$

3. RESULT AND DISCUSSION

Overall, the research results show gamma radiation exposure affects the tire granular characteristics (functional group, density, and crosslink density). Radiation dose and percent granular content of tires also affect the compressive strength and water absorption of the resulting concrete. The research results will be described in detail as follows.

3.1 Tire Granular Characterization

Gamma radiation exposure to polymer (such rubber) may lead to polymer chain scission and crosslinking. Polymer crosslinking can occur through several stages, including the formation of radical species, initiation, propagation, and termination. The degree of polymer chain scission and crosslinking is affected by the radiation dose. In this work, gamma radiation exposure is expected to change the tire granular characteristics. Gamma radiation is expected to increase crosslinking between rubber polymers, so that they have a higher crosslink density. Higher crosslink density leads to increased tire granular hardness. It has the potential to be used as an aggregate as well as sand aggregate. Characteristics changes of before and after irradiated tire granular were identified using functional group, crosslink density, and density. Analysis results of functional group changes of before and after irradiated tire granular are shown in Figure 3.

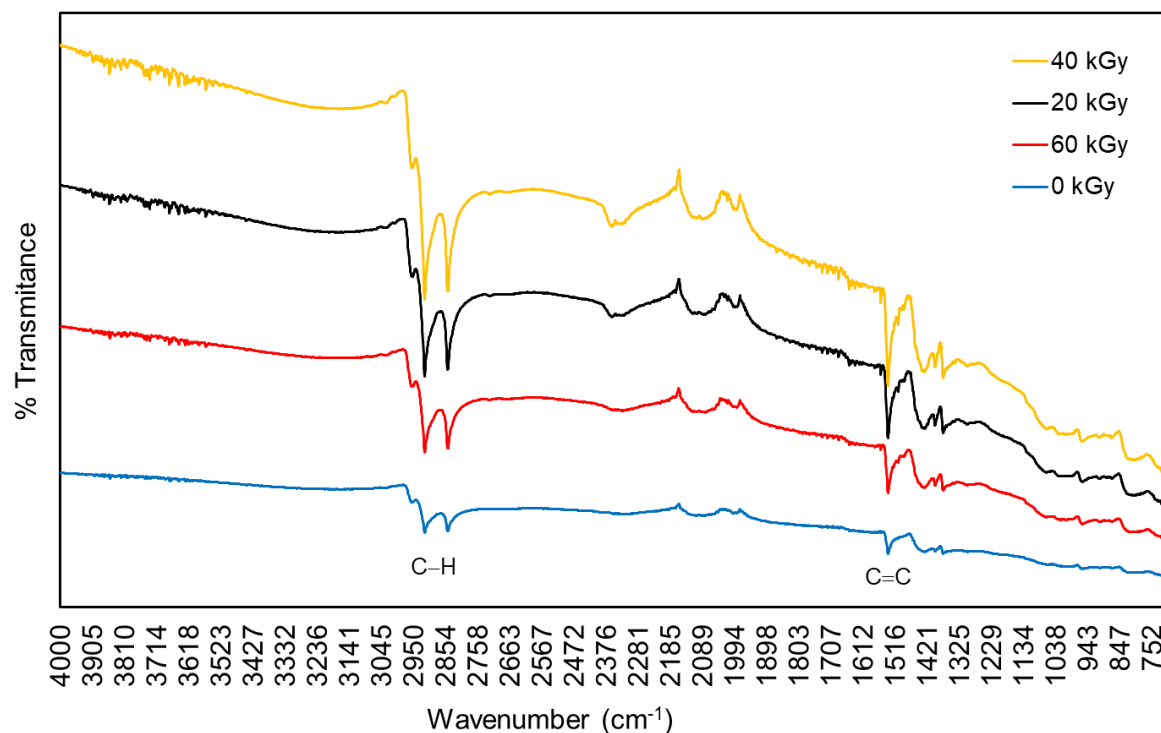


Figure 3. FTIR spectra of tire waste powder

Figure 3 shows the presence of the C–H stretching band of the methylene moieties (CH_2) at the peaks of 2916 cm^{-1} dan 2848 cm^{-1} . A band around 1537 cm^{-1} also appeared, which may be attributed to C=C stretching or the C-S bond in vulcanized rubber [19]. Figure 3 shows the C–H and C=C transmittance spectra of irradiated tire granular is deeper than non-irradiated tire granular (0 kGy). Low transmittance indicates a high population of bonds with vibrational energies that represent the incident infrared light, whereas high transmittance at a frequency indicates that there are few bonds to absorb that infrared light in the sample. This indicates that the C–H and C=C in irradiated tire granular are less than non-irradiated tire granular. C–H reduction is due to the scission of the C and H bonds caused by gamma radiation, thus C atoms form a cross-linked C bond between other rubber polymers and H atoms form H_2 gas. The same situation happened to the C=C group. C=C reduction is due to the scission of the C and C bonds caused by gamma radiation, thus C atoms form a cross-linked C bond between other rubber polymers following the crosslinking scheme presented by previous researchers [10, 11]. It can be inferred that gamma radiation was successful in increasing tire granular crosslinking.

The increase of tire granular crosslinking after irradiation was confirmed by measuring crosslink density as shown in Table 2. Crosslink density is defined as the number of crosslinks per unit volume in a polymer network [18]. Table 2 shows that gamma radiation exposure to 40 kGy caused tire crosslink density to increase ($5.9 \times 10^{-4} \text{ mol/cm}^3$) from non-irradiated tire granular ($4.6 \times 10^{-4} \text{ mol/cm}^3$). This crosslink density increase indicates that there is crosslinking increase in the tire granular due to gamma radiation. However, gamma radiation dose at 60 kGy causes the tire crosslink density to decrease to $2.7 \times 10^{-4} \text{ mol/cm}^3$. It is estimated that the crosslinking scission at 60 kGy as stated by previous researchers. Butyl rubber (the type of rubber in tires) undergoes chain scission when exposed to gamma radiation at a dose of 45 kGy [17]. The tire density measurements in Table 2 also show results that are in line with the value of tire crosslink density.

Table 2. Radiation dose effect to density and crosslink density of tire granular

Radiation Dose (kGy)	Tire Density (g/cm ³)	Tire Crosslink Density (x10 ⁻⁴ mol/cm ³)
0	0.84	4.6
20	0.82	3.4
40	0.86	5.9
60	0.80	2.7

3.2 Concrete Characterization

The concrete quality tested in this study was K70. K70 means the quality of the concrete at 7 days curing. Referring to SNI 03-0349, good quality concrete has a minimum compressive strength of 100 kg/cm² or not worse than K100 and a maximum water absorption of 25%. K100 means the quality of the concrete at 28 days curing [20]. The compressive strength of K100 concrete is calculated using Eq. 4. The effect of radiation dose and percent tire granular content on the compressive strength and water absorption of concrete is described in detail as follows.

$$K100 = \frac{70}{100} \times K70 \quad (4)$$

3.2.1 Radiation Dose Effect

The concrete was cast using the sample C in Table 1, where the tire granular used were irradiated at various doses, including 0, 20, 40 and 60 kGy. The results in Figure 4 show that K70 concrete using tire granular 40 kGy has a higher compressive strength (72.805 kg/cm²) than concrete using tire granular 20 kGy (65.875 kg/cm²) and without irradiation (48.54 kg/cm²). Tire granular modification using gamma radiation to 40 kGy can increase the concrete compressive strength to 150%. Based on the results of the previous tire granular characterization, it is known that radiation dose at 40 kGy can create crosslinks in tire granular which is characterized by an increase in tire crosslink density. The crosslink density causes the tire granular to become harder. This causes the concrete using granular tires of 40 kGy to have a higher compressive strength than tire granular without irradiation.

Figure 4 also shows that K70 concrete using tire granular 60 kGy has a higher compressive strength than concrete using tire granular without irradiation, but its compressive strength is lower than concrete using tire granular 40 kGy. This strength reduction is estimated following the results of the tire granular characterization that have been discussed previously. Tire granular is expected to undergo crosslink scission when exposed to a radiation dose of 60 kGy [17]. The crosslink scission causes the tire crosslink density to decrease so that the tire granular is not harder than the tire granular of 40 kGy. This study conveys earlier studies. Concrete that uses tire granular tires of 50 kGy has a higher compressive strength than concrete that uses tire granular of 100 kGy [14].

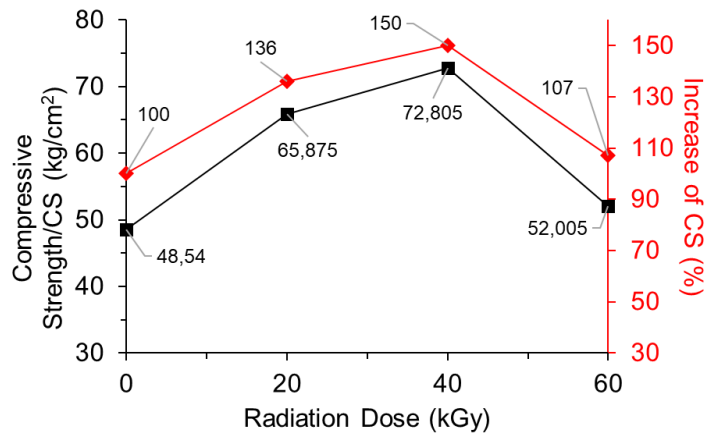


Figure 4. Radiation dose effect to concrete compressive strength at 7 days curing (K70)

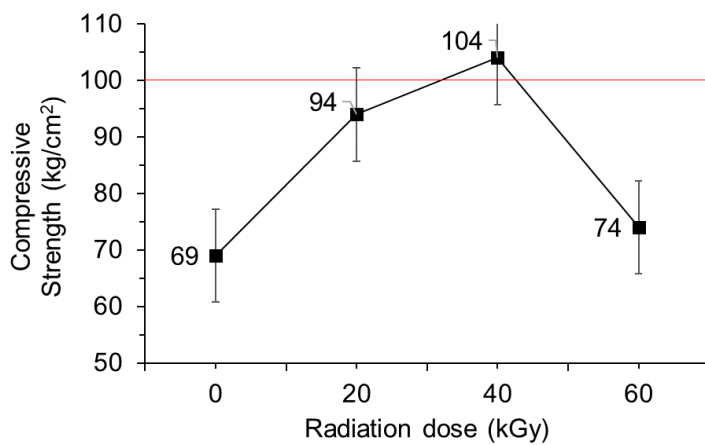


Figure 5. Radiation dose effect to concrete compressive strength at 28 days curing (K100)

Referring to SNI 03-0349 of K100 concrete, only concrete using tire granular of 40 kGy meets the standard (Figure 5). Concrete using tire granular of 20 kGy can also be said to meet the SNI 03-0349 standard when considering the range of standard deviation (error) of the test.

Figure 6 states that each concrete has a water absorption under 25%. Water absorption is the ability of concrete to absorb water when the concrete is hydrated in water until the concrete cannot absorb water anymore because the concrete is saturated. Concrete water absorption is affected by concrete pores or cavities [21]. Referring to SNI 03-0349 of K100 concrete, each concrete that uses tire granular fulfill the standard.

Concrete using tire granular of 40 kGy has the lowest water absorption. According to Table 2, a tire granular of 40 kGy has highest density, so it causes highest concrete density and lowest water absorption. If the sample standard deviation is involved, it might be inferred that the water absorption of concrete is not much different between each concrete (0, 20, 40, and 60 kGy). However, each concrete using tire granular fulfill the SNI 03-0349 standard.

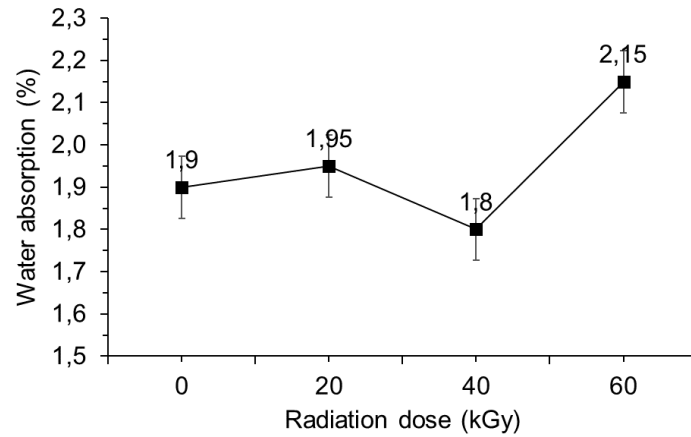


Figure 6. Radiation dose effect to concrete water absorption

3.2.2 Tire Granular Content Effect

The results in Figure 7 show that an increase in the percent tire granular content causes a decrease in the concrete compressive strength. In this work, tire granular serves as a substitute for sand aggregate. According to Table 1, it is known that the increasing use of granular tires, and the decreasing use of sand. This work conveys earlier studies. Concrete using tire granular of 5% has a lower compressive strength than concrete using tire granular of 3% [15]. The compressive strength of concrete decreases as the tire granular content increases [22]. Sand has a higher hardness compared to granular tires. Sand also has a higher density (1.4 g/cm^3) than tire granular (Table 2). It might be inferred concrete E has a lower density than others (Table 3). High-density concrete has a high density and hardness, which causes the concrete to have a high compressive strength.

Based on Figure 7, concrete B and C fulfill the compressive strength standard of SNI 03-0349. The use of tire granular as a substitute for sand aggregates in concrete is a maximum of 4% in order to meet SNI 03-0349 standards.

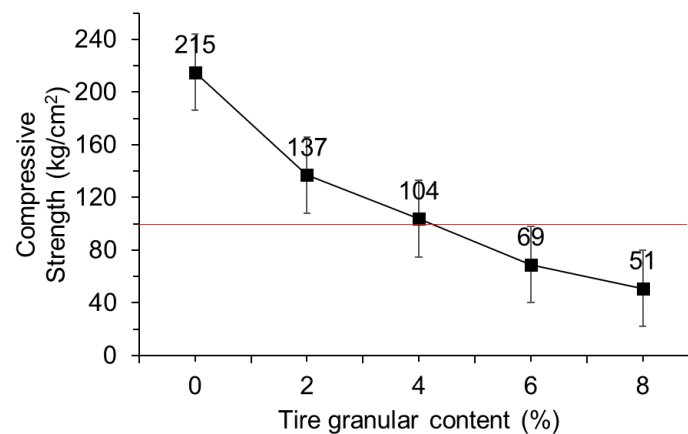
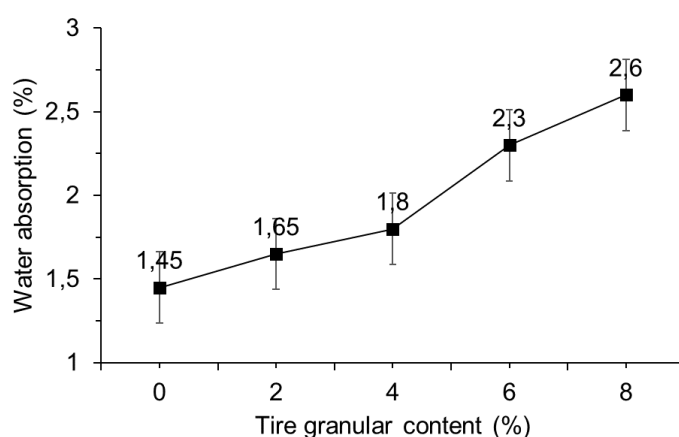


Figure 7. Tire granular content effect to predicted concrete compressive strength at 28 days (K100)

Table 3. Concrete density

Concrete	Tire granular content (%)	Density (g/cm ³)
A	0	2.32
B	2	2.24
C	4	2.22
D	6	2.08
E	8	1.99

The results in Table 3 show that the higher the tire granular content, the lower the density of the concrete. This is because the density of tire granular is lower than sand density. The results of this work conveys earlier studies. Concrete with high tire granular content (10%) has a lower density than concrete with low tire granular content (5%) [8].

**Figure 8. Tire granular content effect to concrete water absorption**

In contrast to the compressive strength, the results in Figure 8 show that the tire granular content is linear to the water absorption. However, the water absorption increase was not statistically significant if according to standard deviation. Figure 8 shows that E concrete has a higher water absorption than other. The water absorption of concrete is affected by the concrete voids [21]. Concrete that has a lot of voids will have a low density. This is supported by the results in Table 3 which shows that E concrete has a lower density than other concretes. However, each concrete in Figure 7 has water absorption under 25%. Referring to SNI 03-0349 of K100 concrete, each concrete still meets the standard.

4. CONCLUSION

Exposing gamma radiation at 40 kGy can increase the crosslink density of tire granular, which indicates polymer crosslinking is successfully achieved. Concrete K100 with irradiated tire granular at 40 kGy (4% w/w) gives compressive strength improvement 150% (104 kg/cm²) compared to non-irradiated tire granular (69 kg/cm²). Even though concrete with irradiated tire granular gives lower compressive strength compared to concrete without irradiated tire granular, concrete with irradiated tire granular optimum content (4%) are still fulfill standard of SNI 03-0349. Concrete with irradiated tire granular content 2%, 4%, 6%, and 8% are also fulfill standard of SNI 03-0349.

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