Gamma-induced grafting of rice straw cellulose with polyvinylpyrrolidone

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Abstract: Cellulose obtained from rice straw was grafted with polyvinylpyrrolidone (Cell-g-PVP). The process was induced by gamma simultaneous irradiation (Co-60). The presence of the grafted PVP chains was confirmed by FT-IR, XRD and EDS methods. The influence of the irradiation dose (20 and 30 kGy) on the reaction efficiency and thermal stability of Cell-g-PVP was investigated. The grafting yield at the dose of 30 kGy was about 56%. The higher dose of radiation resulted in better thermal stability. The AAS results showed that Ni(II), Cu(II), and Cr(II) ions can be adsorbed by Cell-g-PVP. The order of ion metals adsorption capacity of Cell-g-PVP was Cu(II) > Ni(II) > Cr(II). The adsorption isotherm of Cu(II) metal ion using the Cell-g-PVP followed the Langmuir adsorption isotherm whereas the chemical adsorption and the adsorption energy were constant on all sides of the copolymer.

Keywords: cellulose, polyvinylpyrrolidone, gamma irradiation, graft copolymerization.

Indukowane promieniowaniem gamma szczepienie celulozy ze słomy ryżowej poliwinylopirolidonem

Streszczenie: Celulozę otrzymaną ze słomy ryżowej poddano szczepieniu poliwinylopirolidonem (Cell-g-PVP). Proces indukowano promieniowaniem gamma (Co-60). Obecność zaszczepionych łańcuchów PVP potwierdzono metodami FT-IR, XRD i EDS. Zbadano wpływ wielkości dawki promieniowania (20 i 30 kGy) na wydajność reakcji i stabilność termiczną Cell-g-PVP. Stopień zaszczepienia przy dawce 30 kGy wynosił ok. 56%. Większa dawka promieniowania skutkowała większą stabilnością termiczną. Metodą AAS oceniono właściwości adsorpcyjne Cell-g-PVP względem jonów Ni(II), Cu(II) i Cr(II). Zdolność do adsorpcji zmniejszała się w szeregu Cu(II) > Ni(II) > Cr(II). Izoterma adsorpcji jonów Cu(II) miała przebieg zgodny z izotermą adsorpcji Langmuira.

Słowa kluczowe: celuloza, poliwinylopirolidon, promieniowanie gamma, kopolimeryzacja szczepiona.

In recent decades, the rate of industrial development has seriously impacted environmental pollution [1]. Several industrial sectors produce waste in the form of heavy metals, such as metal mining, electroplating, fertilizers, metallurgy, electronics, electroplating, paints and pigments, and battery manufacturing. The known methods have been used to reduce heavy metals conta- minants in wastewater, such as chemical precipitation, coagulation, and flocculation [2], ion exchange, electrochemical oxidation [3], bio-catalytic technology, membrane separation, flotation, and adsorption [4]. Between these methods, adsorption with modified cellulose is the best solution due to its low operating costs, high efficiency, simple operation, and good regeneration [1, 5–8].

Polyvinylpyrrolidone (PVP) is a non-ionic amorphous polymer with C=O, C–N, and –CH$_2$ functional groups [9] that have been widely used in nanoparticle synthesis [10]. The ease of PVP to be solved in water and many other non--aqueous liquids, with the duality of highly polar amide group within the pyrrolidone ring and a polar methylene and methane groups in the ring and along its backbone [11], allow this polymer to be a great stabilizer [12]. Due to these excellent properties, several reports have studied PVP as a composite with cellulose [13–18] or modification with cellulose-based material [19, 20]. Radiation-induced grafting of cotton-cellulose [21] and cellulosic membrane [22] with pyrrolidine monomers has been prepared previously. The copolymer-mixed radiation-induced grafted PVP and acrylamide on cellulose [23] is also reported by our group. We found that the copolymerization with PVP as a major substrate can be enhanced the Cu(II) adsorption. Herein, the direct grafting of polyvinylpyrrolidone (PVP) on cellulose (Cell-g-PVP) by simultaneous irradiation using gamma rays from cobalt-60 is prepared, characterized, and used for applications as an adsorbent for heavy metals such like Cu(II), Ni(II), and Cr(II).

EXPERIMENTAL PART

Materials

The isolated cellulose from rice straw was prepared in Research Center for Radiation Processing Technology, National Research and Innovation Agency (BRIN) laboratory (Jakarta, Indonesia) according to the procedure published elsewhere [24]. Copper sulfate pentahydrate, potassium dichromate, nickel sulfate hexahydrate, and polyvinylpyrrolidone were purchased from Merck (Jakarta, Indonesia). Distilled water was prepared on-site.

Methods

Preparation of cellulose-g-polyvinylpyrrolidone (Cell-g-PVP)

Cellulose (2.70 g) was put in 5 radiation-resistant polypropylene plastic bags. Polyvinylpyrrolidone (PVP) was diluted in distilled water and then added to a cellulose sample with a ratio of 1 : 1 and 0.5 (cellulose: PVP, w/w). Then, it was closed tightly using a vacuum seal machine and irradiated using Co-60 Gamma cell 220 irradiators with irradiation dose variations of 20, 25, 30 and 40 kGy with a dose rate of 2.5 kGy/h. The irradiation was conducted using the simultaneous method in a vacuum polypropylene plastic bag at ambient temperature. The Perspex dosimeter was used to observe the absorbed dose. The grafted sample was washed using distilled water until the filtrate was not slimy, then immersed in the methanol. The filtrate was filtered and dried using filter paper and an oven at a temperature of 70 °C until reaching a constant weight. The degree of grafting was calculated using Equation (1).

$$Grafting\ Degree = \frac{W_t - W_0}{W_0} \cdot 100\% \qquad (1)$$

where: W_0 and W_t are cellulose dry weights before and after grafting (g).

Swelling capacity

Cellulose and synthesized Cell-g-PVP (50 mg) were added to a glass tube with 50 ml of distilled water and allowed to stand for 24 hours at room temperature. The sample is then removed from the solution and any residual water on the sample surface was dried with filter paper. The swelling capacity was calculated using equation (2).

$$Swelling\ capacity\ (\%) = \frac{S_t - S_0}{S_0} \cdot 100\% \qquad (2)$$

where: S_0 and S_t are cellulose dry weight before and after immersing in distilled water (g).

Fourier Transform Infrared Spectroscopy (FT-IR)

The FT-IR spectra with ATR mode of cellulose, cell-g-PVP 20 kGy, and cell-g-PVP 30 kGy were taken with FT-IR instrument (Nicolet iS10, Polymer Chemical Engineering Laboratory of the Polytechnic of STMI Jakarta, Indonesia). The spectra were taken in the wavelength range 400–4,000 cm^{-1} at ambient temperature.

Thermogravimetric analysis (TGA)

Thermogravimetric analysis of cellulose, cell-g-PVP 20 kGy, and cell-g-PVP 30 kGy were carried out with TGA Instruments (TA TGA 55, Polymer Chemical Engineering Laboratory of the Polytechnic of STMI Jakarta, Indonesia). Analyses were performed on 10 mg of samples in a platinum dish programmed with heating sequences at 100°C and held isothermally for 10 minutes. Heating was then continued to 800°C, gradually decreased to 500°C and held for 30 minutes.

Scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDS)

The SEM images and EDS profile of cellulose, cell-g-PVP 20 kGy, and cell-g-PVP 30 kGy were obtained with a SEM-EDS microscope (JEOL-JSM-6510LA, BRIN Kawasan PUSPITEK Serpong, Indonesia). The samples were coated with gold before measurement.

X-ray diffraction (XRD)

X-ray diffraction patterns of cellulose, cell-g-PVP 20 kGy, and cell-g-PVP 30 kGy were obtained with an XRD instrument (PANalytical, EMPYREAN, BRIN Kawasan PUSPITEK Serpong, Indonesia).

Metal ion adsorption analysis with atomic absorption spectroscopy (AAS)

Metal ion adsorption experiments were carried out at various initial concentrations of metal ions. The cell-g-PVP 30 kGy (15 mg) was immersed in a 100 mL solution of metal ions with a concentration variation of 3, 5, 7, and 10 ppm, placed on a shaker at 100 rpm for 3 hours at room temperature, and then filtered using filter paper to take the filtrate. The filtrate was analyzed using the AAS instrument (Analytic Jena ContrAA 800, CTRSM laboratory, BRIN, Indonesia) to determine the final concentration of metal ions. The flame condition used was air-acetylene flame with a blue flame color. The calibration was carried out using a standard solution of metal ions with concentrations of 1, 2, 5, and 10 ppm, respectively, before analyzing the filtrate. Adsorption performance was evaluated as adsorption capacity following Equation 3.

$$Adsorption\ capacity\ (\%) = \frac{C_0 - C_t}{C_0} \cdot 100\% \qquad (3)$$

where: C_0 and C_t are concentrations of metal ions in solution (mg/L).

The absorbance results of Cell-g-PVP 30 kGy for application of Cu(II) solution were then used to determine adsorption isothermal. The adsorption isothermal was calculated based on Equation 4 for Langmuir isotherms or Equation 5 for Freundlich isotherms.

$$\frac{C_e}{q_e} = \frac{1}{Q_0 K_l} + \frac{1}{Q_0} \cdot C_e \qquad (4)$$

where: C_e is concentration at equilibrium (mg/L); q_e is the number of adsorbed molecules on the surface of the adsorbent (mg/g); Q_0 is adsorption capacity (mg/g) and K_l is Langmuir isotherm constant.

$$\log q_e = \log K_f + \frac{1}{n} \log C_e \qquad (5)$$

where: C_e is concentration at equilibrium (mg/L); q_e is the number of adsorbed molecules on the surface of the adsorbent (mg/g); n = adsorption intensity and K_f is Freundlich isotherm constant.
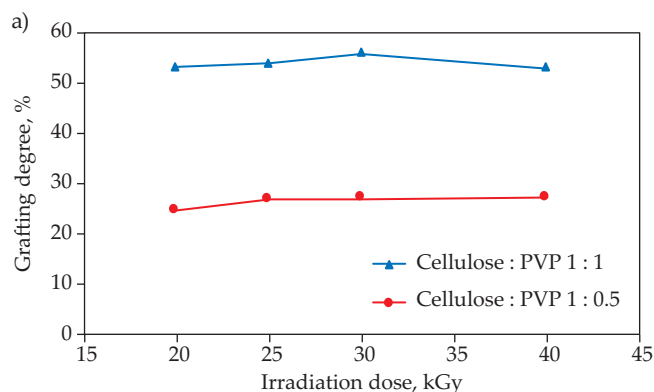
RESULTS AND DISCUSSION

The effect of irradiation dose on the grafting yield

Figure 1a shows the relationship between the degree of grafting and the irradiation dose of the Cell-g-PVP copolymer. When the dose was increased from 20 to 30 kGy, the number of free radicals formed might have massively occurred in the cellulose skeleton which is indicated by the increased percentage of grafted PVP. As for the dose of 40 kGy, the grafting yield is decreased due to the number of free radicals that can initiate the scissions which induce degradation instead of copolymerization. Both ratios cellulose and PVP 1:1 and 1:0.5 showed a similar tendency of the possible PVP grafted on cellulose based on almost twice the amount of grafting degree. Hence, cellulose and PVP ratio of 1:1 with irradiation of 30 kGy which resulted in 55.8% grafting yield is preferable to synthesized Cell-g-PVP. The difference between the irradiation dose and the absorbed dose is less than 3%.

Determination of swelling capacity in water

As a representative of its application as an ion metals adsorbent, the Cell-g-PVP swelling capacity is carried out to measure the number of water molecules that are absorbed into the sample. The swelling capacity was determined by weighing the mass of dry cellulose and the mass of cellulose after immersion in water. Based on the results of a study on various irradiation doses used were 20 and 30 kGy (Fig. 1b), the value of swelling capacity was 346%, 224%, and 248% respectively.

Chemical structure

To confirm the presence of cellulose grafted PVP (Cell-g-PVP), Fourier transform infrared (FT-IR) spectroscopy was performed on the cellulose before and after the grafting. The FT-IR spectra of cellulose and Cell-g-PVP at the irradiation doses of 20 and 30 kGy are presented in Fig. 2. The FT-IR spectrum of ungrafted cellulose showed
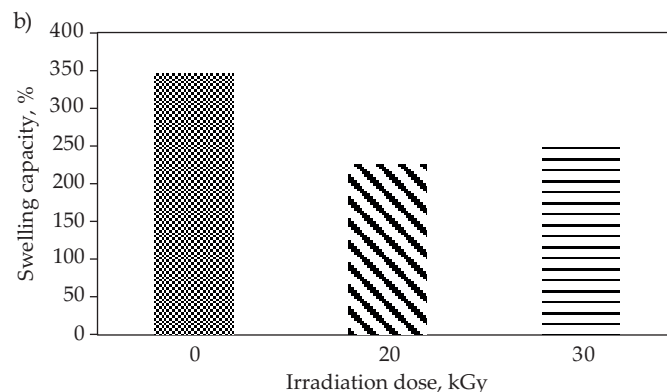

Fig. 1. The relationship between the effect of irradiation dose on grafting with cellulose and PVP ratio of 1:1 and 1:0.5, respectively (a) and the relationship between irradiation dose to swelling capacity in Cell-g-PVP graft copolymers (b)
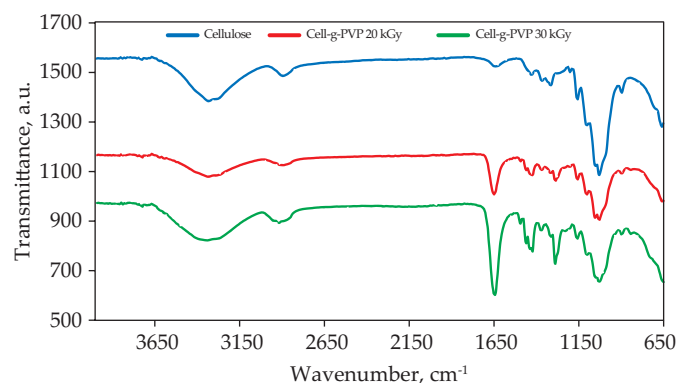
Fig. 2. FT-IR spectra of cellulose, Cell-g-PVP copolymers

peaks at 3333 cm[-1] (-OH stretch), 2896 cm[-1] (C-H stretch), 1640 cm[-1] (-OH bend), 1428 cm[-1] (C-H strain) and range 1158–1315 cm[-1] (stretched CO). A similar peak for cellulose can be clearly identified in the FT-IR spectra of Cell-g-PVP 20 kGy and Cell-g-PVP 30 kGy. Specific peak at 3335 cm[-1] (N-H group), 1650 cm[-1] (C = O stretch), 1315 cm[-1] (C-H bending of the methylene group), 1289 cm[-1] (CN stretch), and 896 cm[-1] (vibration bending = CH), were shown in the FT-IR spectra of Cell-g-PVP 20 kGy and Cell-g-PVP 30 kGy, which indicates the presence of a PVP-derived functional group [22], which proves the grafting. In the case of Cell-g-PVP 30 kGy, a bigger peak intensity was found at 1650 cm[-1], which indicates a higher efficiency of the grafting.

Thermal stability

Thermal stability of nongrafted and grafted cellulose was conducted using thermogravimetric analysis (TGA). The results were presented in Fig. 3 and summarized in Table 1. Based on the temperature at which decomposition occurred with a 5% weight loss (Table 1), the degradation of pure cellulose began at 253°C, while of Cell-g-PVP samples occurred at 50–60°C. This indicates an increase in the organic content of grafted cellulose due to the incorporation of a low molecular weight PVP compound. The temperature at which 20% weight loss occurs for pure cellulose, Cell-g-PVP 20 kGy and Cell-g-PVP 30 kGy were around 320°C what was related
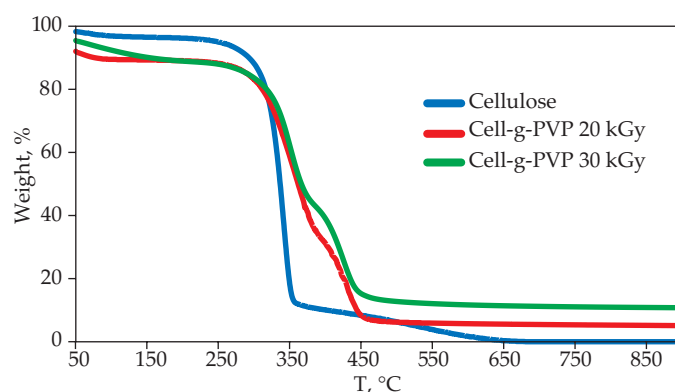
T a b l e 1. TGA data of cellulose and Cell-g-PVP copolymers

Sample	$T_{5\%}$ °C	$T_{20\%}$ °C	$T_{50\%}$ °C	Residue at 530°C, %
Cellulose	253	319	339	5.4
Cell-g-PVP 20 kGy	50	314	364	6.1
Cell-g-PVP 30 kGy	61	320	370	12.9

$T_{5\%}$, $T_{20\%}$, $T_{50\%}$ – the decomposition temperature of 5, 20 and 50% weight loss, respectively.

to cellulose degradation [25]. The slight improvement of the copolymer thermal stability can be seen at decomposition temperature for 50% weight loss of Cell-g-PVP 20 kGy and Cell-g-PVP 30 kGy whereas the decomposition has occurred relatively higher in comparison with the pure cellulose. The slight improvement in thermal stability of grafted cellulose is evidenced by the significantly higher of 50% weight loss temperature compared to neat cellulose. The residue at 530 °C of Cell-g-PVP 20 kGy and Cell-g-PVP 30 kGy also can present slightly improved thermal stability after the PVP is grafted into the cellulose.

X-ray diffraction characterization

The X-ray diffraction (XRD) patterns of cellulose derived from rice husk, Cell-g-PVP 20 kGy, and Cell-g-PVP 30 kGy are illustrated in Fig. 4. The diffraction characteristic peaks of cellulose from rice husk located at 14.44°, 15.88°, 19.60°, 21.720°, 33.77° corresponded to the (16), (13), (20), (100), and (10) planes, respectively. The characteristic peaks of the Cell-g-PVP 20 kGy were located at 15.14°, 16.37°, 20.23°, 21.869°, and 34.16° which corresponded to the planes of (34), (10), (13), (100), and (9). Moreover, the diffraction characteristic peaks of Cell-g-PVP 30 kGy were located at 15.1°, 16.5°, 20.33°, 22.36°, and 34.00° that was corresponding to the (25), (21), (11), (100), and (12) planes, respectively. The XRD patterns of PVP are known as not showing any distinctive peaks due to their amorphous nature [26]. In correlation, the Cell-g-PVP 20 kGy
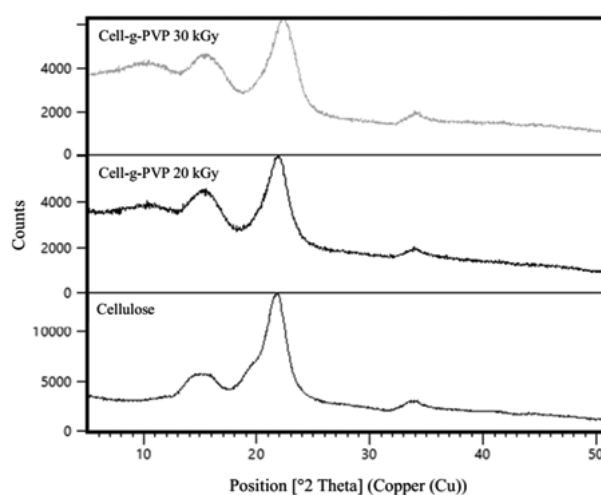

Fig. 3. TGA curves of cellulose and Cell-g-PVP copolymers

Fig. 4. X-ray diffraction pattern of cellulose and Cell-g-PVP copolymers
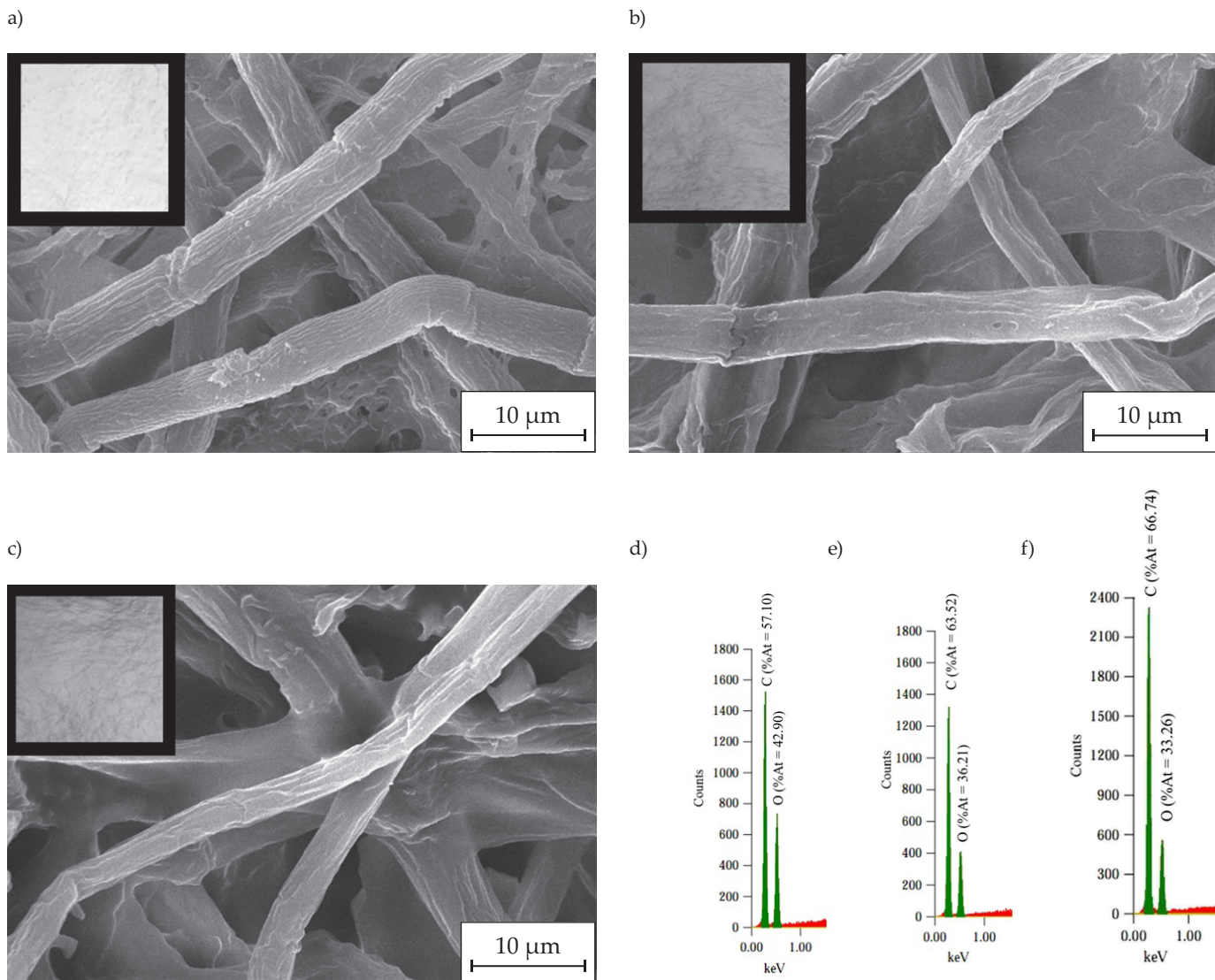
a)

b)

c)

d)   e)   f)

Fig. 5. SEM images: a) cellulose, b) Cell-g-PVP 20 kGy, c) Cell-g-PVP 30 kGy. EDS profile: d) cellulose, e) Cell-g-PVP 20 kGy, f) Cell--g-PVP 30 kGy

and Cell-g-PVP 30 kGy patterns had distinct diffraction peaks of cellulose and PVP; however, the increase in the peak intensity cannot be clearly identified compared to those analyzed by FT-IR.

Morphology and elemental analysis with SEM-EDS

The SEM images (Fig. 5a–c) show the pure cellulose and Cell-g-PVP copolymers morphology as microfibrils with a diameter of less than 10 μm. The Cell-g-PVP 20 kGy (Fig. 5b) and Cell-g-PVP 30 kGy (Fig. 5c) microfibrils tend to be narrower with the rough surfaces due to the PVP introduction in comparison with cellulose (Fig. 5a). The ratio of C and O elements of cellulose, Cell--g-PVP 20 kGy and Cell-g-PVP 30 kGy is 1.33, 1.75, and 2.01, respectively, based on EDS analysis (Fig. 5d–f) in which indicating the increment of PVP grafting in cellulose surface. These results are coherent with the successful grafted PVP into cellulose analysis with FT-IR (Fig. 2) and XRD (Fig. 4).

Analysis of adsorption capacity in various concentration and metal ions

The determination of the adsorption capacity of Cell-g-PVP 20 kGy and Cell-g-PVP 30 kGy was conducted with an adsorbent mass of 15 mg and observed with 3 ppm, 5 ppm, 7 ppm, and 10 ppm of 3 different metal ions–Cu(II), Ni(II) and Cr(II). In the process, the adsorption was carried out using a shaker at room temperature to avoid the effect of temperature ion movement. According to Figure 6, the adsorption capacity of Cell-g-PVP 20 kGy that contacted with a low concentration of metal ions was relatively lower than Cell-g-PVP 30 kGy. These results are correlated with the higher number of grafted PVP on cellulose surfaces due to the high dose of irradiation (Fig. 1a) which can be associated with the pyrrolidone fragment structure possibility of making the tautomeric forms [16]. The adsorption capacities for Cell-g-PVP 30 kGy were 56%, 32%, and 12% for 10 ppm of Cu(II), Ni(II), and Cr(II), respectively. Based on these numbers,
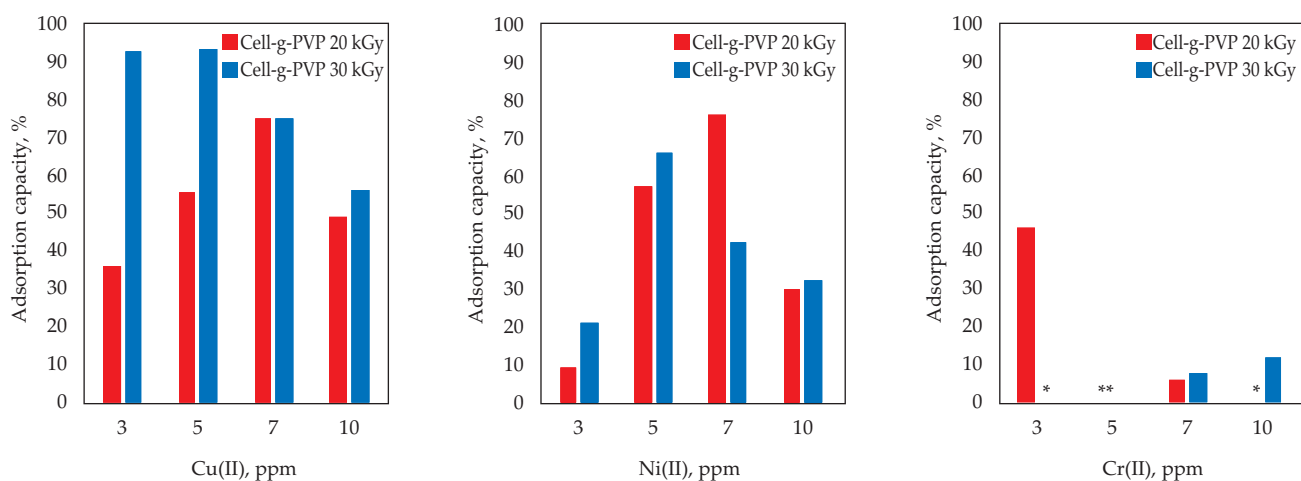
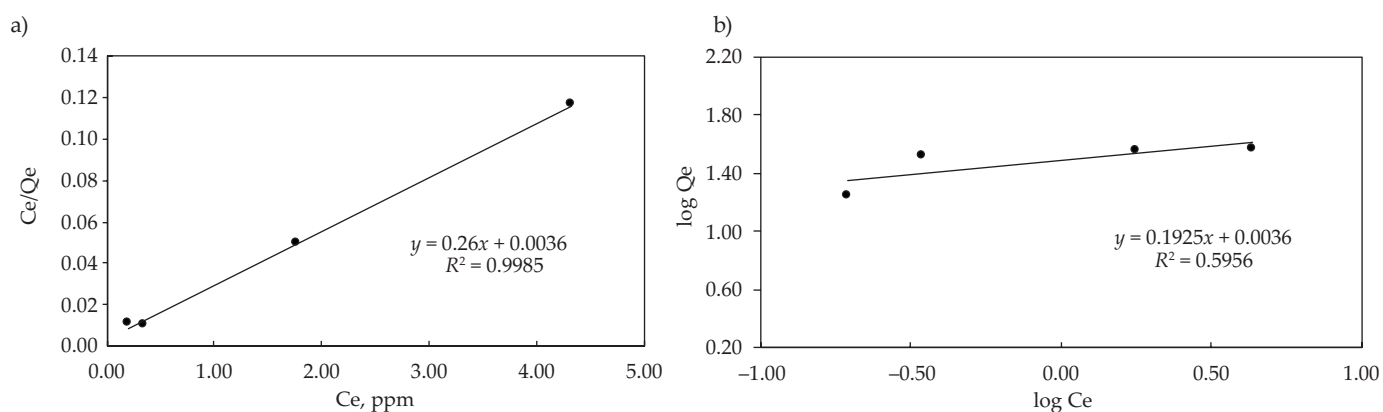**Fig.6. Effect of Cu(II), Ni(II), and Cr(II) concentrations on Cell-g-PVP copolymers adsorption capacity**

Fig. 7. Cu(II) Langmuir (a) and Freundlich isotherm adsorption (b) for Cell-g-PVP30 kGy copolymer

the order of Cell--g-PVP 30 kGy as adsorbent of ion metal is Cu > Ni > Cr. Impressively, these results showed more high efficiency for simple preparation of Cu(II) adsorbent rather that the mixed-copolymer of cellulose with PVP and acrylamide [23]. The introduction of PVP solution in cellulose adsorbent via temperature modification of 30°C and pH 6 condition is known to be dramatically improved the sorption capacity of Cu(II) [18].

The adsorption phenomena are illustrated by the adsorption isotherm which has a correlation between the equilibrium concentration and the amount of adsorbate absorbed by the adsorbent weight unity. Langmuir (homogeneous adsorption) and Freundlich (heterogeneous adsorption) isotherm are used in liquid-solid adsorption. Linear line equations between Ce to Ce/Qe were created when performing analysis using the Langmuir model. In contrast, Log Ce to Log Qe line was created when performing analysis using the Freundlich model. The analysis results can be seen in Fig. 7a and b. The regression results for the Langmuir and Freundlich isotherm equation are 0.998 and 0.595 respectively (Fig. 7a and b). Thus, it concluded that the Cell-g-PVP 30 kGy adsorption process at all different concentrations follows the Langmuir isotherm model because it has higher linearity than the Freundlich isotherm model. From the Langmuir isotherm equation, the valid model of monomolecular adsorption onto a sorbent with energetically equivalent adsorption centers [20], it is assumed that the adsorbent can adsorb with a homogeneous surface and constant adsorption energy on all sides of the surface. The Cell-g-PVP 30 kGy adsorbent does meet this condition. The Langmuir isotherm equation obtained is following as $y = 0.026x + 0.003$ based on Equation 4. The Kl and Qmax values are 8.97 and 38.46 mg/g, respectively. According to the calculations, the maximum adsorption capacity of Cell-g-PVP 30 kGy adsorbent against Cu(II) is 38.46 mg/g.

CONCLUSIONS

The optimal irradiation dose of PVP grafting onto cellulose (Cell-g-PVP) was achieved at 30 kGy. The order of optimum adsorption capacity of Cell-g-PVP adsorbent in metal ions adsorption is Cu > Ni > Cr. The Cu(II) adsorption isotherm follows the Langmuir adsorption isotherm model with a linearity value of 0.998. The multi-functionalized cellulose with radiation-induced grafting may be a promising biomaterial for waste water with ion metals contaminants in the future.

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