U(VI) Adsorption onto Low Dose Radiation Acclimated *Tradescantia Fluminensis*

Jun Liu (✉ edwardnuclear_liu@163.com)

Chengdu University of Technology

Junxiang Shu

Chengdu University of Technology

Shilong Shi

Chengdu University of Technology

Chao Li

Chengdu University of Technology

Bing Li

Chengdu University of Technology

Dabiao Liang

Chengdu University of Technology

Fei Li

Chengdu University of Technology

Tu Lan

Sichuan University

Jiali Liao

Sichuan University

Ning Liu

Sichuan University

Kunming Zhang

Xinjiang Uygur Autonomous Region Research Institute of Measurement and Testing

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Research Article

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Abstract

The plants that long-term grown in radiation area could be acclimated by low dose radiation (LDR). In this paper, LDR acclimated *Tradescantia fluminensis* (Commelinaceae) was first collected as a biomass adsorbent towards uranium adsorption. Comparative experiments verified the potential radiation effects of LDR acclimation, moreover, q_e values of the wild and LDR acclimated *Tradescantia fluminensis* were ~16 mg/g and 20 mg/g, respectively. U(VI) adsorption amount gradually reached equilibrium after 180 min shaking, and the adsorption process described well by the pseudo-second-order model. However, ionic strength has no obvious effect on the q_e values. SEM and TG-DSC suggested good structural stability of LDR acclimated *Tradescantia fluminensis* during the U(VI) adsorption process. FTIR and XPS verified the surface coordination of U(VI) via -OH and -COOH groups on LDR acclimated *Tradescantia fluminensis* surface, and the increasing amounts of the two groups could account for the improved U(VI) adsorption capacity compared to the wild *Tradescantia fluminensis*. Our present work can indicate that LDR acclimation could be a novel way to obtain biomass adsorbents for U(VI) removal from aqueous solutions.

1. Introduction

Hexavalent uranium can easily migrate in the environment, resulting in many cases of uranium contamination to the pedosphere and hydrosphere. Several tens of mg/L uranium could be measured around the waste disposal sites, especially in some acid mine water (Li et al. 2015b). Besides, the migration and enrichment of U(VI) in human body through the food chain could cause severe toxic injury and radiation damage to the kidneys, livers and other organs. For human health and ecological security protection reasons, uranium removal from contaminated nuclear industrial effluent makes a lot of sense.

Adsorption takes advantages of low cost, operation simplicity, eco-friendly technology and high wastewater treatment efficiency (de Freitas et al. 2019). Many biomass adsorbents, such as coir pith (Parab et al. 2005), *Trichoderma harzianum* (Akhtar et al. 2007), *Rhizopus arrhizus* (Wang et al. 2010), *Arthrobacter* (Carvajal et al. 2012), tea wastes (Li et al. 2015a), *Saccharomyces cerevisiae* (Zheng et al. 2018), marine fungus (Han et al. 2020) etc. have been tested. For example, Aytas et al. (Aytas et al. 2011) found that algae and yeast immobilized on silica gel could improve interaction properties between the bi-functionalized biocomposite and U(VI) ions. Bayramoglu et al. (Bayramoglu et al. 2015) improved U(VI) adsorption capacity via the synthesis and application of polyethyleneimine and amidoxime modified *Spirulina platensis*. The binding functional groups such as carboxylate, hydroxyl, amidoxime and others on live, dead or modified biomass surface play a key role in coordinating U(VI) in solutions (Wang et al. 2015). However, the biomass modification methods mainly focused on chemical modification, researches on radiation effects on biomass mainly aimed at the assessment of enzymatic activity or production of microorganisms. Potential radiation effects, especially for plants are rarely studied.

Highly energetic particles produced by radiation could destroy the cell structure or induce the chemical reactions to attack cell. The major difference between radiation damages of animals and plants' cells is that the former is fatal, whereas plants display excellent cell turnover and regeneration property. Plants growing in nuclear radiation exclusion zone may also have the repair mechanism of DNA protection. Early reports

(Amiro & Sheppard 1994) found that even the dose rates reached up to 65 mGy/h, the herbaceous plant community still thrived. Low dose radiation (LDR) has a certain stimulation on plants include the dormancy breaking, germination promotion, acceleration of growth and development, rooting induction, increase of yield and disease resistance improvement, etc (Zaka et al. 2004). Besides, the meristem in plants is believed to have sensitivity to radiation, and the mechanism may be involved in the repairing enzymes activity caused by the biological organism injury. But as far as we know, no studies reported on U(VI) adsorption onto plants that are growing in radiation area. Moreover, studies on U(VI) adsorption onto these unique biological samples will be conducive to deducing the potential effects of LDR acclimation on U(VI) adsorption efficiency by biomass adsorbents.

Tradescantia fluminensis is a perennial evergreen herb that grown in warm and humid climatic conditions, native to central Brazil, Uruguay and Paraguay and belong to commelinaceae. Herein, U(VI) adsorption efficiency was first determined for both wild and LDR acclimated *Tradescantia fluminensis* samples that collected from radiation area in Southwest China. The potential effects of LDR acclimation were deduced, and the key factors such as contact time, pH, ionic strength and U(VI) concentration affected on U(VI) adsorption had been conducted. In addition, U(VI) loaded samples were also characterized by FTIR, SEM-EDS, TG-DSC, and XPS for purpose of the possible reactive mechanisms discussion.

2. Materials And Methods

2.1 Chemicals

1 g/L uranium stock solution was prepared based on our published research (Liu et al. 2017). All the used chemicals were analytical grade.

2.2 Characterization

Thermo Nicolet 6700 spectrophotometer (USA) was used for recording the FTIR to identify the surface functional groups. Surface morphology and EDS analyses were conducted using a ZEISS SUPRA 40 (Germany) and X-Max (UK). A TGA/DSC2 apparatus (Mettler-Toledo, Switzerland) carried out the TG-DSC data acquisition. The electronic structure information was obtained by an Axis-Ultra, Kratos (UK).

2.3 Collection of *Tradescantia fluminensis* biomass

Tradescantia fluminensis biomass was collected from the radiation area (Located in the grassland of Applied Nuclear Techniques in Geosciences Key Laboratory of Sichuan, Chengdu University of Technology, Sichuan, China) that contains the Th-232 isotope radioactive source over a period of 10 years. Two kinds of *Tradescantia fluminensis* biomass were tested, i.e. the wild *Tradescantia fluminensis* growing in the natural background radiation environment ($< 214 \pm 37.4$ nGy/h) and the LDR acclimated *Tradescantia fluminensis* growing in the radiation environment that exceeding the maximum background limits ($> 214 \pm 37.4$ nGy/h) (Fig. 1). Two kinds of *Tradescantia fluminensis* biomass samples are randomly and equally collected from the range of the above two area, respectively.

2.4 Pretreatment of *Tradescantia fluminensis* biomass adsorbents

Silt and sand on the fresh and sliced biomass (with roots) were dried at 90°C for 24 h after rinsing with running water. Then transferred it to a mortar and grind to powder with a 200-mesh sieve. The treated biomass was sealed and stored in a desiccator as use.

2.5 U(VI)determination

U(VI) concentration was measured on a UV-2450 UV-VIS spectrophotometer (SHIMADZU, Japan) following the Arsenazo-III spectrophotometric method (Liu et al. 2016a). And the only improvement is that the percent content of Arsenazo-III decreased from 0.1–0.06% for economic considerations.

2.6 Adsorption experiments

Adsorption of U(VI) by *Tradescantia fluminensis* was conducted using static equilibrium method. 0.01 g *Tradescantia fluminensis* was added into 25 mL U(VI) working solutions with various concentrations after pH adjustment. The mixture was continuously shaken at 25°C until the adsorption equilibrium was achieved. Uranium concentrations in the supernatant liquid were determined via the mentioned above method at 651.8 nm. The adsorption capacity q_e (mg/g) was calculated as follows:

$$q_e = (C_0 - C_e) \times V / m \quad (1)$$

where C_0 and C_e refer to the initial and equilibrium U(VI) concentrations (mg/L), respectively, V is the solution volume (L), and m is the biomass adsorbent mass (g).

3. Results And Discussion

3.1 Effect of impact factors

Kinetics of the adsorption process could be determined by the study of time effect. Clearly, during the first 15 min, U(VI) adsorption amount increased rapidly then reached almost saturation at 180 min (Fig. 2A). The ultimate uranium adsorption capacity of the wild and LDR acclimated *Tradescantia fluminensis* were ~15.2 mg/g and ~19.0 mg/g, respectively.

U(VI) adsorption on the wild and LDR acclimated *Tradescantia fluminensis* as a function of initial pH value was shown in Fig. 2B. The adsorption amount increased linearly from 0.5 and 2.0 mg/g to 15.9 and 19.3 mg/g with an increase of pH from 1.5 to 4.5 for the wild and LDR acclimated *Tradescantia fluminensis*. When the solution pH values are too low, the competitive adsorption between H^+ and UO_2^{2+} on *Tradescantia fluminensis* surface is too severe due to the high H^+ content, resulting in the small q_e values. Besides, the hydroxyl and carboxyl groups on *Tradescantia fluminensis* surface are easily protonated at low solution pH values. The repulsive electrostatic interactions with H^+ caused the poor UO_2^{2+} adsorption performance. As

the pH values increased, the decreasing competition and repulsive electrostatic interactions contributed the increasing of U(VI) adsorption capacity. The upper limit pH value was set at 4.5 in order to avoid the formation of $\text{UO}_2(\text{OH})_2 \cdot \text{H}_2\text{O}$ precipitate (Liu et al. 2016b). Moreover, the uranium adsorption capacity improved for LDR acclimated *Tradescantia fluminensis*. Bank et al. (Bank et al. 2008) compared the clay mineral reactivity of γ -irradiated sediments and non-sterile samples, but the former sorbed significantly more U(VI) because γ -induced reduction decreased the surface charge of iron-bearing minerals. Liu et al. (Liu et al. 2015) indicated that cellulose, treated by high absorbed dose irradiation in oxygen, formed -C=O and -COOH groups because of the oxidative degradation. Accordingly, uranium adsorption capacity of LDR acclimated *Tradescantia fluminensis* enhanced because of the higher amounts of the carboxyl groups existed on adsorbent's surface compared to the wild one.

The competition between cations and UO_2^{2+} for the active sites may also control the adsorption process. Herein, NaNO_3 , a key electrolyte in nuclear wastewater, was selected as the affecting factor, and the results are displayed in Fig. 2C. One can observe a good tolerance of *Tradescantia fluminensis* even the Na^+ concentration reached 1.0 mol/L, which provided selectivity response to *Tradescantia fluminensis* for UO_2^{2+} against Na^+ . Saini and Melo (Saini & Melo 2013) indicated that the strong affinity between UO_2^{2+} and melanin binding sites contributes to the little impact of Na^+ and NO_3^- on uranium adsorption by melanin. Similarly, no significantly effect of NaNO_3 ions in wastewater on uranium adsorption by modified *Cystoseira sp.* biomasses was observed in Gök et al.'s work (Gök et al. 2017). The observed trend for the ionic strength effect could be an advantage for the usability of the LDR acclimated *Tradescantia fluminensis* in real uranium containing wastewater conditions.

3.2 Adsorption kinetics and isotherms

Adsorption kinetics study aimed at evaluation of adsorption efficiency and mechanism controlling. Herein, different kinetic models, i.e. the pseudo-first-order model and the pseudo-second-order model were fitted. Eqs. (2) and (3) expressed the the linear form:

$$\ln(q_e - q_t) = \ln q_e - k_1 t$$

2

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t$$

3

where q_t is the U(VI) adsorbed amount (mg/g) at any time t , k_1 (min^{-1}) and k_2 (mg/g min^{-1}) are the pseudo-first-order and pseudo-second-order rate constants. The fitting results and the relevant kinetics parameters are plotted and listed in Fig. 3 and Table 1, respectively.

A higher correlation coefficient (R^2) and a value of calculated q_{e2} that is close to $q_{e,exp}$ based on pseudo-second-order model indicates that adsorption process may be involved in sharing or exchanging of electrons between the adsorbent and adsorbate, in other words, chemisorption dominates the rate-limiting step of the

reaction. Gül et al. (Gül et al. 2019) suggested that UO_2^{2+} adsorption on lichen mainly through the electrostatic interactions and the interaction of the carboxyl groups. Khani (Khani 2011) indicated that uranium removal by *Padina* sp. algae biomass could be associated with ion exchange mechanism. Considering the abundant functional groups, such as -COOH and -OH existed on *Tradescantia fluminensis* surface, they could provide surface active sites for uranium adsorption. Furthermore, the q_e values of the LDR acclimated *Tradescantia fluminensis* was always higher than that of the wild *Tradescantia fluminensis*, indicating the potential improvement U(VI) adsorption capacity of *Tradescantia fluminensis* affected by LDR acclimation.

Bhat et al. (Bhat et al. 2008) found that uranium adsorption curve data of *Catenella repens* (a red alga) could be well described by both the linearized Langmuir and Freundlich adsorption isotherms at pH 4.5. Therefore, adsorption isotherms had also been evaluated (Fig. 7 and Table 2).

The equilibrium data was evaluated by the Langmuir and Freundlich models, which could be given by linear Eq. (4) and (5):

$$\frac{C_e}{q_e} = \frac{1}{K_L q_m} + \frac{C_e}{q_m}$$

4

$$\ln q_e = \ln K_F + \frac{1}{n_F} \ln C_e$$

5

where q_m (mg/g) is the theoretical maximum adsorption capacity, K_L (L/mg), K_F (mg/g (L/mg) $^{1/n}$) and n_F are the Langmuir and Freundlich constants that related to the adsorption energy, adsorption capacity and adsorption intensity, respectively.

The relatively low R^2 values of Langmuir models in our work may be associated with the mixed of dried plant organs, such as roots, stems and leaves in *Tradescantia fluminensis* samples, resulting the non-monolayer and heterogeneous adsorption process. Note that the correlation coefficient of Freundlich models is high and Eq. (4) is an empirical equation, a multilayer adsorption on a heterogeneous surface may dominate the present adsorption process. Besides, n_F value greater than 1 demonstrated the favorable nature of adsorption on *Tradescantia fluminensis* biomass (Cheira et al. 2020).

Table 1 Kinetic parameters for U(VI) adsorption onto the wild and LDR acclimated *Tradescantia fluminensis*.

Samples	$q_{e, exp}$ (mg·g ⁻¹)	Pseudo-first-order kinetics			Pseudo-second-order kinetics		
		q_{e1} (mg·g ⁻¹)	k_1 (min ⁻¹)	R^2	q_{e2} (mg·g ⁻¹)	k_2 (g·mg ⁻¹ ·min ⁻¹)	R^2
LDR acclimated <i>Tradescantia fluminensis</i>	~19.0	~12.5	0.0128	0.913	~20.6	0.00259	0.997
Wild <i>Tradescantia fluminensis</i>	~15.2	~8.53	0.0126	0.850	~16.3	0.00427	0.998

Table 2 Isotherms parameters for U(VI) adsorption by the wild and LDR acclimated *Tradescantia fluminensis*.

Samples	Langmuir isotherm model			Freundlich isotherm model		
	q_m (mg/g)	K_L (L/mg)	R^2	n_F	K_F	R^2
LDR acclimated <i>Tradescantia fluminensis</i>	~556	0.000864	0.102	1.25	1.18	0.976
Wild <i>Tradescantia fluminensis</i>	~213	0.00308	0.547	1.07	0.598	0.975

3.3 FTIR

Figure 4 showed the FTIR spectra of the wild and LDR acclimated *Tradescantia fluminensis* before and after U(VI) adsorption. The wild *Tradescantia fluminensis* samples have obvious absorption bands in the whole wavenumber range due to the existence of the cellulose, hemicellulose, carbohydrates, etc. (Figure 4a). The hydroxyl stretching vibration at ~3426 cm⁻¹ indicated that -OH existed on the surface of *Tradescantia fluminensis* (Tewatia et al. 2021). The C-H stretching vibration of -CH₃ (methyl) and -CH₂ (methylene) in the carbon chain was observed at ~2920 and 2852 cm⁻¹. The C=O stretching vibration of the carboxyl group corresponded to the sharp band at 1626 cm⁻¹. The bands appeared at 1384 cm⁻¹ demonstrated the presence of C-OH (Cui et al. 2015). The -OH in cellulose could be verified at around 1243 cm⁻¹. And the characteristic bands of cellulose could also be observed at 1106 and 1074 cm⁻¹. The C-H bending vibration in aromatic nucleus appeared at 832.72 and 779.94 cm⁻¹. It can be seen that the structure added no significant new chemical groups, whereas the -COOH and C-OH at 1626 and 1384 cm⁻¹ enhanced after LDR acclimation, indicating that the increase of the -COOH and -OH on the LDR acclimated *Tradescantia fluminensis* surface (Figure 4b). Xu et al. (Xu et al. 2007) and Akram et al. (Akram et al. 2012) found that γ -irradiation wouldn't change the group structures of konjac glucomannan and mushroom samples based on

the FT-IR spectra. Moreover, Sun et al. (Sun et al. 2013) manifested that the increasing absorbed dose enhanced the characteristic peak ascribed to the carbonyl groups. Evidence from the irradiation in acacia and karaya gum suggested that the absorbance of the functional groups like –OH, –COOH, uronic acid and pyranose units also increased (Hamdani et al. 2017).

After U(VI) adsorption, the band at 1626 and 1384 cm^{-1} that related to –COOH and –OH gradually weakened (Fig. 4c and d). Besides, a new IR absorption bands correlated with the O-U-O stretching for U(VI) appeared at 925 and 917 cm^{-1} (Jain et al. 2018). These may be acceptable evidence for the surface complexation between carboxyl and hydroxyl groups and U(VI) occurring on *Tradescantia fluminensis* surface.

3.4 SEM-EDS

The surface morphologies and element types could be observed and detected via the SEM and EDS analyzes. Obviously, C and O are the mainly elements on the plant surface, and the morphologies of leaves and stems with plant fibers displayed clearly on the *Tradescantia fluminensis* surface (Fig. 5). Actually, any visible physical effect caused by irradiation may not be presented on the biomass (Heredia-Guerrero et al. 2012, Oneh Abu et al. 2006). The stems showed folded surface, which was in line with the isothermal models results, indicating the heterogeneous uranium adsorption process. The similar microstructure and characteristic peaks suggested good structural stability of *Tradescantia fluminensis* during the U(VI) adsorption process.

3.5 TG-DSC

The TG-DSC curves of the wild and LDR acclimated *Tradescantia fluminensis* before and after U load were shown in Fig. 6. As shown, two stages, associated with approximate 15.0~20% (room temperature to ~100°C) and 46~48% (100°C to 500°C) weight loss in *Tradescantia fluminensis*, could be divided. The loss of the moisture and low organic content of the samples and the pyrolysis of organic matter such as cellulose, hemicellulose and lignin could be the main reasons (Müsellim et al. 2018). For all heating rates, an exothermic behavior could be found in the reactions on the basis of DSC plots.

3.6 XPS

The bonding environment and surface chemical composition of the wild and LDR acclimated *Tradescantia fluminensis* before and after U load were analyzed through XPS. U4f peak, presented in wide scan data (Fig. 7a) on U load sample, was the clear evidence for the U adsorption ability of *Tradescantia fluminensis*. Further analysis of valence state from the fitting curves of U4f spectrum high-resolution XPS spectra (Fig. 7b) revealed the existence of hexavalent uranium because doublet U4f_{7/2} and U4f_{5/2} peaks presented at ~382.3 eV and ~393.1 eV.

The O1s XPS spectra of four samples can be deconvoluted into two peaks at 531.4±0.2 and 532.7 ±0.1 eV, corresponding to C=O and C-O (hydroxyl) bond (Ahmed et al. 2021). No significant differences were observed between the wild and LDR acclimated *Tradescantia fluminensis*. Ansón-Casaos et al. (Ansón-Casaos et al. 2014) also found that the O1s spectra upon γ -irradiation for few-layered graphene materials displayed few

variations, and they seem to occur quite randomly. However, the normalized intensity of the hydroxyl increased for both the U(VI)-loaded *Tradescantia fluminensis*.

Three peaks can be observed in C1s XPS spectra for *Tradescantia fluminensis* samples, as depicted in Fig. 7d. The peaks at binding energy of ~ 284.8 , 286.3 ± 0.2 and 288.2 ± 0.2 eV can be related to the intensity of C=C, C-O (hydroxyl) and C=O. Increasing irradiation dose weakened the relative intensity of the C-O (hydroxyl) and C=O bands. Wan et al. (Wan et al. 2005) also noted that the amount of -OH in C1s region of carbon fibers decreased after gamma irradiation. Besides, the hydroxyl normalized intensity also increased for both U(VI)-loaded *Tradescantia fluminensis*, which is in line with the results of O1s.

3.7 Probable adsorption mechanisms

On the basis of FTIR and XPS spectra, the amounts of carboxyls and hydroxyls on the surface of the wild *Tradescantia fluminensis* increased after LDR acclimation, accompanying by the -OH and -COOH groups coordination for U(VI). Moreover, multilayer chemical adsorption on a heterogeneous surface was described well for the U(VI) adsorption by both the wild and LDR acclimated *Tradescantia fluminensis* according to the isotherm and kinetics analyses. In general, the complexation of carboxyls and hydroxyls on the surface of the LDR acclimated *Tradescantia fluminensis* played a key role in UO_2^{2+} adsorption. Besides, the results of batch adsorption experiments correlated well with the characterization data. The probable mechanisms of U(VI) adsorption on the wild and LDR acclimated *Tradescantia fluminensis* were schematically shown in Fig. 8.

4. Conclusions

In summary, we first report a plant, i.e. *Tradescantia fluminensis* collected from the radiation area for adsorption of uranium. Due to the potential LDR acclimation effect, the uranium adsorption capacity of *Tradescantia fluminensis* improved, and the q_e values of the wild and LDR acclimated *Tradescantia fluminensis* were about 16 mg/g and 20 mg/g when $\text{pH}_{\text{initial}}$ was 4.5 and C_0 was 50 mg/L. Adsorption kinetics demonstrated a multilayer adsorption on *Tradescantia fluminensis* surface. Higher pH and initial uranium concentration benefited the U(VI) adsorption process, whereas ionic strength has no obvious effect on the q_e values. Characterization results suggested that the carboxyl and hydroxyl groups on *Tradescantia fluminensis* surface may complex U(VI), and the LDR acclimated *Tradescantia fluminensis* contained a larger number of the two groups than the wild ones. Besides, good structural stability of the LDR acclimated plant remained during the U(VI) adsorption process. In summary, the LDR acclimated *Tradescantia fluminensis* may be a novel candidate for radioactive wastewater uranium removal.

Declarations

Availability of data and materials Not applicable

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Authors contributions Jun Liu: Conceptualization, Supervision, Writing-original draft, Funding acquisition

Junxiang Shu: Investigation, Formal analysis

Shilong Shi: Conceptualization, Writing-original draft, Methodology

Chao Li: Methodology, Formal analysis

Bing Li: Investigation, Formal analysis

Dabiao Liang: Investigation, Formal analysis

Fei Li: Conceptualization, Validation, Methodology

Tu Lan: Writing-review & editing

Jiali Liao: Writing-review & editing

Ning Liu: Supervision, Writing-review & editing

Kunming Zhang: Validation, Formal analysis

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Figures

Figure 1

The distribution map of dose rate in radiation area.

Figure 2

Effect of contact time (A), initial pH (B), ion strength (C) and initial U(VI) concentration (D) on U(VI) adsorption by the wild and LDR acclimated *Tradescantia fluminensis*.

Figure 3

Plots for (A) Pseudo-first-order, (B) Pseudo-second-order kinetics (C) Langmuir model and (D) Freundlich model fitting curves.

Figure 4

The FTIR spectra of the (a) wild, (b) LDR acclimated, (c) wild U(VI) loaded and (d) LDR acclimated U(VI) loaded *Tradescantia fluminensis*.

Figure 5

The SEM images and EDS analysis of the wild and LDR acclimated *Tradescantia fluminensis* before and after U(VI) adsorption.

Figure 6

TG-DSC curves of the wild and LDR acclimated *Tradescantia fluminensis* before and after U(VI) adsorption.

Figure 7

(a) Typical XPS survey spectrum and the high resolution spectra of (b) U4f, (c) O1s and (d) C1s for the wild and LDR acclimated *Tradescantia fluminensis* before and after U(VI) adsorption.

Figure 8

Possible mechanisms for U(VI) adsorption on the wild and LDR acclimated *Tradescantia fluminensis*.