Modeling of Innovation Research Clusters in the Field of Radioactive Waste Utilization

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Abstract. The situation with the processing and disposal of radioactive waste, including the liquid phase, is not resolved. Many questions remain regarding the environmental safety of processes and the technological implementation of the most energy-efficient solutions. Thus, the article's main attention is paid to theoretical studies of the development innovation directions of radioactive waste processing using the VOSviewer software tool. The clusters were formed under modeling directions of application of radiolysis for radioactive waste utilization: the red cluster includes research of radionuclide sorption processes and their concentration for radiolysis of liquid media; the green cluster concerns factors of influence on water radiolysis in hydrogen production; the yellow cluster includes research areas of natural processes related to radiolysis; the blue cluster is connected with mathematical modeling of radiolysis process with fuel production and engineering implementation and the purple cluster related to the processes of radioactive waste management and disposal as well as the application of radiolysis for this purpose. The cluster simulations of nanoparticle and radiolysis applications for radioactive waste treatment have resulted in a new energy recovery strategy. The development of new matrix materials in combination with nanoparticles for the agglomeration and concentration of radionuclides is a promising innovation method improving radiolysis under hydrogen production from radioactive waste.

Keywords: radioactive waste, nanoparticle, radiolysis, cluster simulations, software tool.

1 Introduction

Responding to the growing demand for sustainable development requires using the world's resources as productively and minimally as possible and keeping their negative impact on human health and the environment as low as possible.

The study [1] found that wind and nuclear power plants have the highest sustainability performance. Fuel elements using hydrogen from coal and natural gas have been identified as the most unfavorable conversion technologies in terms of sustainability.

However, the efficient and safe operation of the nuclear power plant (NPP) is very topical today. Thus, radioactive contamination of natural areas is one of the longest-lasting anthropogenic impacts on the environment. Duarte et al. (2019) have analyzed the de novo transcriptome of the Scottish pine population growing in the Chernobyl affected area, which is still contaminated with radionuclides due to the 1986 nuclear accident. Transcriptome profiles indicate a clear picture of the adaptive stress response, which seems to depend on the dose.

These adaptive reactions, which are caused by radiation doses 30 times lower than those recognized as safe for biota species under international standards, suggest that environmental management for radiation protection should be reviewed [2].

The disaster at the Japanese Fukushima-1 NPP in March 2011 is the largest radiation accident in the world after the Chernobyl nuclear power plant. Awareness of the causes of the incident and the scale of this disaster's consequences allows us to draw useful lessons for the future and develop a balanced attitude to the further development of nuclear energy, considering risks to human life and health.

The total release of radionuclides of iodine and cesium at Fukushima-1 so far corresponds to 10 % of what was thrown out during the Chernobyl accident (excluding uranium and plutonium that were raised during a fire from the Chernobyl reactor core). The destroyed Fukushima-1 reactors' total capacity is almost four times higher than the capacity of the 4th Chernobyl nuclear power plant unit. To this must be added the spent fuel assemblies (FA) that have

accumulated over the 40 years of operation of the Fukushima-1, which also played a negative role in developing the accident [3].

Besides, a significant amount of radioactive waste is generated at existing nuclear power plants. In general, they are generated through a wide range of activities related to nuclear facilities' operation, the use of closed radioactive sources in the industry, the production of mineral fertilizers, etc. (Azarov et al., 2018). For example, in the fuel elements of nuclear reactors, radioactive isotopes of xenon and krypton are produced significantly. One has to reckon with this during the regeneration of nuclear fuel. Naturally, dealing with radioactive gas is very inconvenient. For this purpose, use the γ-radiation of spent fuel elements of nuclear reactors and other sources, as well as radiation sources. Methyl alcohol for synthesis may contain up to 30 % water. Along with ethylene glycol, formaldehyde, water, carbon monoxide, hydrogen, and methane are formed [3].

Thus, improving the sorption ability of known types of materials is still relevant, and there is a high demand for new composite materials. Primarily they have high disinfection efficiency for solid materials, including the soil complex, and are also suitable for the elimination of the harmful effects associated with radiation pollution of ecosystems.

2 Literature Review

The radioactivity level in the waste affects the selection of its different management options due to its shielding requirements. The current internationally accepted classification system is based on the activity level and half-life. This system classifies the radioactive wastes to exempt (EW), low- and intermediate-level wastes (LILW), which may be subdivided into short-lived (LILW-SL) and long-lived (LILW-LL) wastes, and high-level wastes (HLW). Treatment is an essential phase in the management of radioactive waste. It aims to reduce the volume of generated wastes to enhance safety and/or reduce the costs of further management phases. After the treatment phase, the wastes split into two portions. The first is a small volume of concentrate that contains the bulk of radionuclides kept in the management system. The second is a large volume portion with low radioactivity that allows its discharge to the environment after meeting the regulatory requirements. To facilitate liquid radioactive waste management, they were categorized into aqueous and organic liquid wastes [4].

The solution to the safe treatment of radioactive waste is carried out by creating a multi-barrier system for their isolation from the sphere of human life and environmental protection. In this case, the population's effective dose due to radioactive waste after their disposal should not exceed 10 μSv / year.

Studies of radiolytic hydrogen sources were conducted in Depending on the dispersibility of the spent nuclear material, the concentration of soluble forms of radionuclides, the degree of vaporization of the bulk materials, and water quality, calculation of the hydrogen yield rate was performed [5].

According to the State Agency of Ukraine on Exclusion Zone Management, a liquid radioactive waste treatment plant has been launched at the decommissioned Chornobyl nuclear power plant's site in Kyiv region. The liquid radioactive waste treatment plant has begun work at the Chornobyl NPP. According to Chornobyl NPP specialists, liquid waste is processed in several stages and, as a result, it is solidified in the form of a cement compound, which is a safer form of radioactive waste storage and disposal [6].

However, the situation with the processing and disposal of radioactive waste, including the liquid phase, is not resolved. Many questions remain regarding the environmental safety of processes and the technological implementation of the most energy-efficient solutions.

In this regard, the possibility of using the radiolysis of water and new matrix materials is of interest.

The trend in studies on the fixation of radionuclides with nanoparticles in the Scopus database was analyzed (Fig. 1). It should be noted that there is a growing interest in this area of research, which can be attributed to the widespread introduction of nanotechnology in various applications.
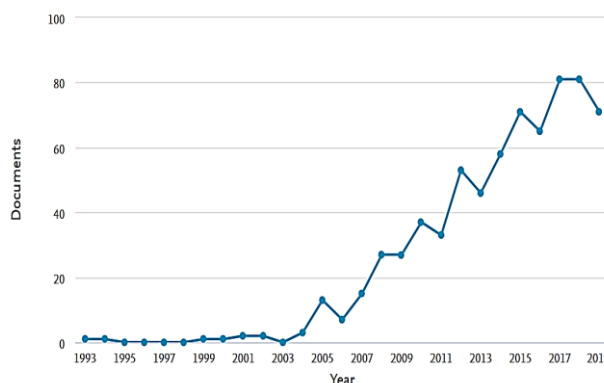

Figure 1 – A sampling of documents by year according to keywords "nanoparticle" and "radionuclide" in the Scopus database

Various combinations of keywords in the initial search for publications in the Scopus database were used for the analysis. In the initial search, the emphasis was on radiolysis and radioactive waste. It should be noted that the dynamics of the study of the subject matter in this area varies considerably by years (Fig. 2).

The article's main attention is paid to theoretical studies of the development innovation directions of radioactive waste processing using the VOS viewer software tool. To achieve the aim, the following tasks were set:
- modeling trends of nanoparticles usage in the field of radioactive waste management with the allocation of the main clusters behind the keywords in publishing activity;
- modeling trends of application of radiolysis in radioactive waste utilization with the allocation of the

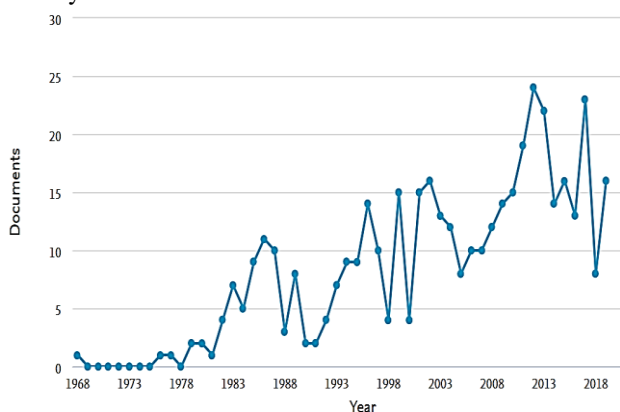main clusters behind the keywords in the publication activity.

Figure 2 – A sampling of documents by year according to keywords "radioactive wastes" and "radiolysis" in the Scopus database

3 Research Methodology

The data on publication activity from the Scopus science database were used for modeling. The tools of the VOS viewer software product for visualization of clusters were used. When forming the General network visualization map by default, the program identifies several large zones (clusters), in which the interrelationships of keywords separate research areas in the study area. The greater is the number of neighboring elements and the smaller is the distance between these elements and a point of interest, the higher is the element density. Overlay visualization has been chosen as a more effective tool for checking the latest trends in time scale studies. The size of circles corresponds to the predominance of terms when publishing studies in this area. The distribution of colors depends on the year of publication (average for a cluster), with the last ones being in yellow. The main core clusters are determined by the timing of their relationships with each other.

4 Results and Discussion

Primary water radiolysis products include several chemical species: e_{aq}^-, HO·, H·, HO$_2$·, H$_3$O$^+$, OH$^-$, H$_2$O$_2$ and H$_2$ [7]. The main reactions are occurring during the three stages of water radiolysis (Table 1).

Table 1 – Different stages and their products in gamma radiation radiolysis of aqueous media (Abedini A. et al, 2016)

Different stages of radiolysis of water	Important reactions
Physical stage ($<10^{-15}$ s)	$H_2O \rightarrow H_2O^+ + e^-$ $H_2O \rightarrow H_2O^*$
Physico-chemical stage ($\sim 10^{-15} - 10^{-12}$ s)	$H_2O^+ + H_2O \rightarrow OH + H_3O$ $H_2O^* \rightarrow H_2O$ $e^- \rightarrow e_{th}^- \rightarrow e_{tr}^- \rightarrow e_{aq}^-$
Non-homogeneous chemical stage ($10^{-12} - 10^{-6}$ s)	$H_2O \rightarrow e_{aq}^-$, H, OH, H$_2$, H$_2O_2$, H$^+$, OH$^-$

Where H$_2$O$^+$ is the ionized water molecule, H$_2$O* is the exited water molecule, and sub-excitations electrons e_{th}^- and e_{tr}^- are thermalized and trapped electrons, respectively [8]. Recently, in water-cooled nuclear reactors, radiolysis processes in water (in liquid and vapor states) and a vapor-metal reaction are considered a source of molecular hydrogen. The release of molecular hydrogen characterizes the radiolytic processes of hydrogen accumulation in reactors. They were observed during homogeneous radiolysis of water, which did not consider the effects of radiation and radiation-thermal processes of structural materials in contact with water on the accumulation of molecular hydrogen [9].

The radiolysis of water can be strongly affected by solid/liquid interfaces. In heterogeneous systems, all three stages can be modified from those taking place in bulk water. The energy deposition is different and various energy transfer processes can occur between the initial ionizing radiation-matter interaction and the chemical events observed in water. These phenomena can correspond to a change in the system's dose or to a change in initial radiolytic yields. Moreover, the reaction and diffusion of species can be modified by the presence of a solid phase during the chemical stage. All these phenomena are particularly salient when nanoporous materials with large specific surfaces are used (more than 50 m^2/g), as specific interfacial phenomena can thus be revealed. The respective energy levels in water and in silica are presented in Figure 3 [7].
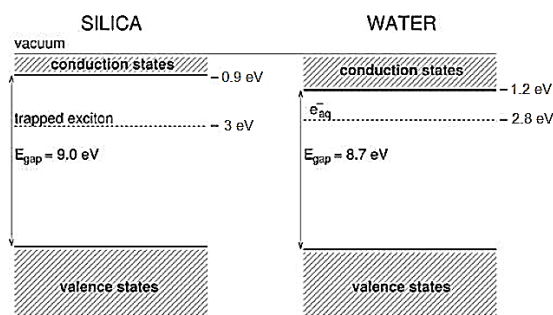

Figure 3 – Scheme of energy levels in water and silica. The conduction band edges concerning the vacuum level are taken as V$_0$(H$_2$O) = −1.2 eV and V$_0$(SiO$_2$) = −0.9 eV, respectively. The difference between the valence and conduction band edge is 9.0 eV in silica and 8.7 eV in water (Le Caër S., 2011)

The density of spurs along a radiation track is an important parameter in determining radiolysis products' chemical yields. For the low linear energy transfer (LET) radiation, the inelastic collision means the free path of the radiation (the primary electron) liquid water at 25 °C is about 1 μm. At the same time, the spur size is about 20 nm [10].

Gamma rays have a smaller LET than charged particles. Consequently, their penetrating distance in a matrix is greater than α and β particles. The greater penetrating distance of γ radiation complicates modeling the radiolytic γ flux because possible curvature of the solid–water interface needs to be considered. Curved interfaces are often relevant for radiolysis studies.

The irradiance for each particle is determined by the angle of incidence (δ) and the inverse square of the distance traveled (R) (Fig. 4) [11].
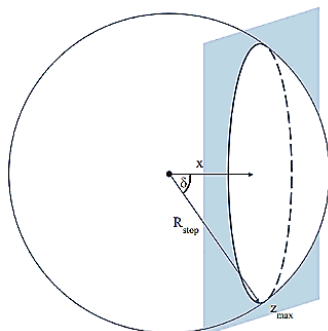

Figure 4 – Schematic illustration for the α and β equations, depicting the path of isotropic radiation. The gray planar surface represents the "solid–water" interface (water to the right of the plane) (Dzaugis Mary E. et al., 2015)

R_{stop} is the stopping distance of the traveling α or β particle, x is the distance from the interface where the radionuclide is located, and δ is the angle of incidence (the angle between particle's path and the normal to the planar interface) [11].

The parameters regulating such energy transfer include oxide band gap, energy migration distance, water adsorption, etc. However, the theoretical justification of the mechanisms of processes and phenomena occurring during the radiolysis of water in heterogeneous media requires further study and establishment. Thus, the chemistry induced by ionizing radiation at the oxide/adsorbate interface has many practical applications (e. g., transformation or decay of pollutants). Therefore, there is a need to fundamentally understand the radiolytic processes occurring on oxide surfaces [7].

Figures 5, 6 present the visualization by clusters of the main modern trends in publishing activity behind the research topic.
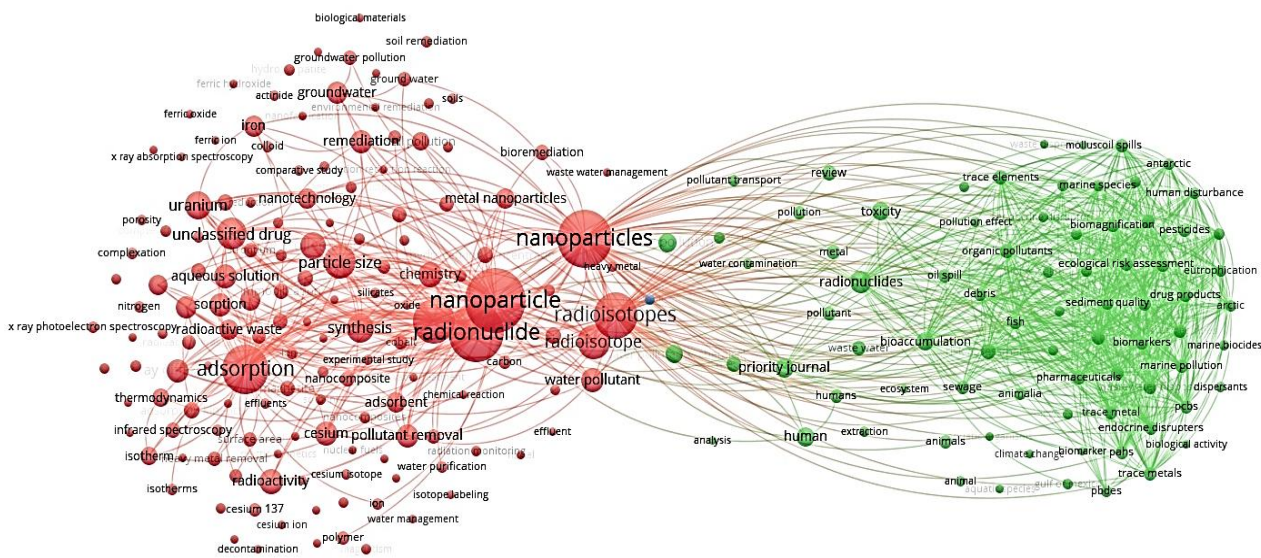

Figure 5 – General network visualization under nanoparticles usage in the field of radioactive waste management (constructed via VOSViewer v.1.6.15): 3 clusters, 12454 links, 35267 total link strength
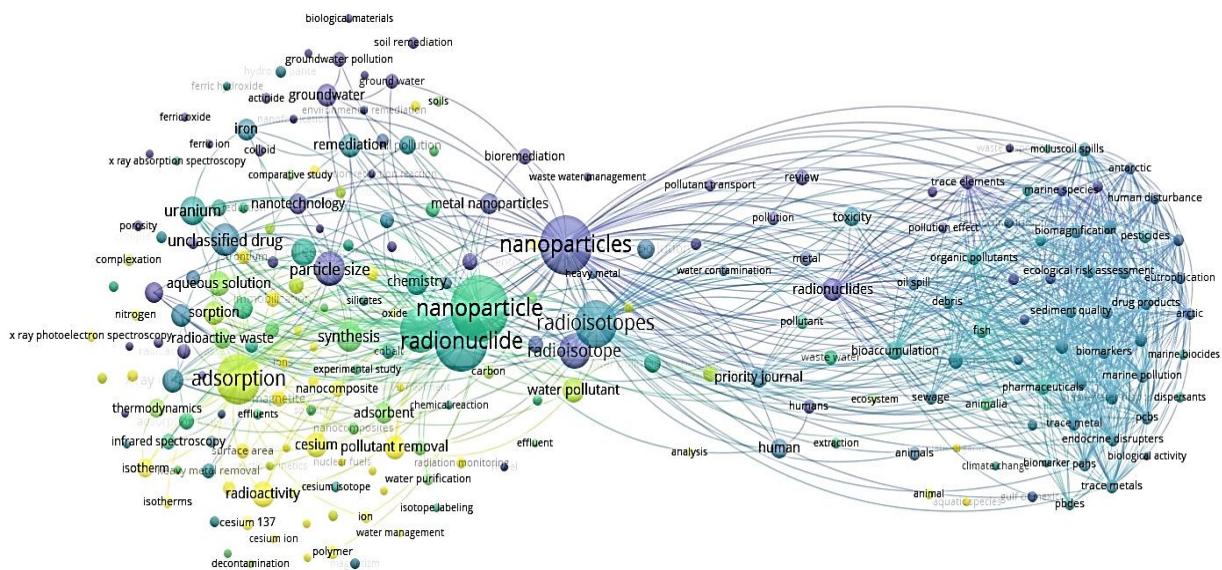

Figure 6 – Overlay visualization of most frequent terms under nanoparticles usage in the field of radioactive waste management (constructed via VOSViewer v.1.6.15), where the score of the item is the time since publication

The clusters were formed: a red cluster – investigation of sorption processes for removal of radioactive elements from liquid media [12–14] green cluster is related to the assessment of environmental and economic risks of radioactive contamination, bioaccumulation processes in the ecosystem and the impact on the human body (medicine application), while considering the possibility of applying nanotechnology to reduce these impacts [15–17]. The blue cluster is also distinguished, but it is still at the formation stage, so in essence, its selection is not clear. It is in the field of research of the red cluster.

Radiolytically synthesized nanoparticles are in the form of colloidal particles, which possess a huge surface-to-volume ratio and high specific surface area. As a result, a large part of the particle atoms' surface can be in contact with the surrounding liquid. This implies the formation of soluble macromolecules, which increase the rate of interactions or fasten the reactions. Thus, colloidal nanoparticles are thermo-dynamically unstable, which in the absence of counteracting force, will grow, and a colloidal system with nanoparticles in various shapes will be formed [18].

Payne et al (2013) considers a solution for decontamination of solid materials and preventing the spread of radioactive materials from a contaminated surface, which consists in the development of colloidal-stable nanosized selective sorbents that can penetrate (in the liquid phase) through contaminated solid materials and, in addition, easily separated by filtration and flocculation by polymer flocculants. Polymer nanoparticles (size range 50–500 nm) bearing carboxyl and epoxy groups on the surface will be synthesized by free radical polymerization [19].

In the study by Norrfors et al. (2016), bentonite was investigated, which is positioned as a radionuclide adsorbent. The adsorption of Th (IV), U (VI), Np (V), Tc (VII), and Pu (IV) on fractionated fractions of montmorillonite in synthetic carbonated groundwaters was studied. Montmorillonite colloids obtained by fractionation of crude clay material in the presence of organic matter at the initial stage of separation, there is a significant decrease in the absorption of Th and Pu [20].

In addition, the study (Runde W. et al, 2002) presented the results of investigated the absorption of Np and Pu on inorganic colloidal particles in water. The solubilities of plutonium, determined experimentally at pH 6.0, 7.0, and 8.5, are approximately two orders of magnitude higher than those calculated using the existing thermodynamic database indicating the effect of colloidal Pu (IV) species (Fig. 7) [21].

Despite real radioactive wastes containing different isotopes of varying concentrations, most tested sorbent materials were conducted using single and bi-solute competitive adsorption. There is a need to study these sorbents' behavior for removing the potential radionuclides as a multicomponent system [22].
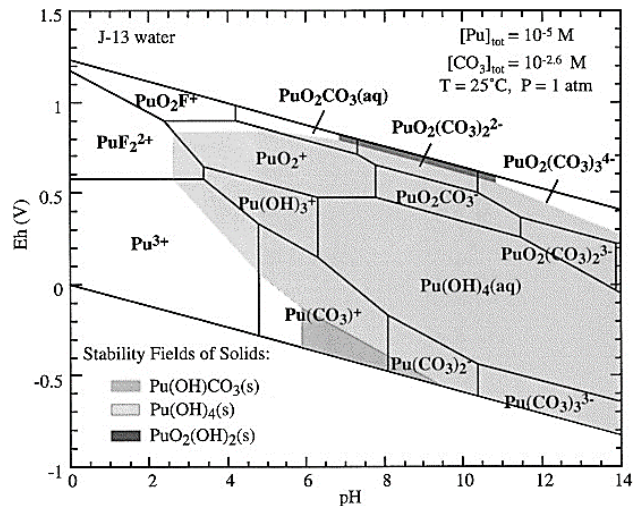

Figure 7 – Field formation of thermodynamically stable compounds with radionuclides (for example, plutonium) (Runde W. et al., 2002)

In the study by Pipíška et al. (2020) investigated pyrogenic carbon materials obtained as a result of the pyrolysis process of various raw material wastes, e. g., thermo-chemically transformed from wood chips, corn cobs, green garden waste, cherry stones, precipitation of municipal wastewater, etc. at slow pyrolysis in the atmosphere of N^2. Experiments on cesium desorption adsorption were carried out using radioisotope ^{137}Cs. Although the tested materials differ significantly in removing $^{+}$Cs, the absorption of Cs ions can be explained by Freundlich's surface adsorption mechanisms. The highest maximum sorption capacity was achieved for pine cones, chips, and garden waste of pyrogenic materials and reached 95.9–126 µmol g. These results demonstrate the suitability of selected pyrogenic carbonaceous materials as Cs adsorbents, potentially suitable as reaction barriers for contaminated wastewaters containing Cs [23].

Preliminary or post-synthetic modification of sorbent materials is also intensively studied to increase their overall stability, tunability, and sorption capacity without changing or damaging the basic matrix structure [24].

In the study by Satoshi Seino et al. (2001) was reported on experiments employing TiO_2 and Al_2O_3 nanoparticle materials performed under various conditions, e.g., changing total absorbed dose, dose rate, amount of the nanoparticles, nanoparticle's size and processing procedure of nanoparticles. These are discussed to clarify the mechanism of enhancement of the hydrogen yield. It was indicated that reactions enhancing the hydrogen evolution are activated both by the γ-ray irradiation and existence of nanoparticles, and that surface of nanoparticles are deeply involved. Hydrogen yield depended on agglomerated particle size rather than that of the primary particle and chemical species of nanoparticles. The enhancement mechanism is dominated possibly by the radiolysis process rather than the photocatalytic process [25].

Dzaugis et al. (2015) used the model to calculate the radiolytic production distribution in the water surrounding spent nuclear fuel. This study was chosen this example because it illustrates the applicability of the method to calculate water radiolysis by γ radiation at a curved interface. The result has important implications for safe handling, disposal, and storage of spent nuclear fuel [11]. From the water irradiated with γ-rays, hydrogen gas generates by radiolysis. In the case when the oxide nanoparticles such as SiO_2, TiO_2, Al_2O_3, or ZrO_2 disperse in the water, the enhancement of the hydrogen gas generation has been observed. However, the cause of the enhancement is not obvious so far. This would be since the irradiation effects on the water, and the nanoparticles have not been distinguished. In the study by Kojima et al. (2006) SiO_2 nanoparticles have been pre-irradiated with [60]Co γ-rays before the experiments of radiolysis of water with the particles dispersing in it irradiation with [60]Co γ-rays. The present work results seem to show that the hydrogen gas generation in the particle dispersing water strongly depends on some changes in the characteristics of the particles induced by γ-ray pre-irradiation [26].

The prospects of using as a carrier of sorption-active compounds glass-crystalline glysoalumosilicate microspheres formed during high-temperature combustion of pulverized coal at thermal power plants are studied. The presence of an amorphous aluminosilicate component with the content from 15 to 62 wt. % in different fractions gives the possibility to synthesize zeolite phases with ion-exchange and sorption properties on the surface of the zeolite phases pherospheres [27].

The heterogeneous radiolysis of water was performed under the static conditions in special ampules. The kinetics of accumulation of molecular hydrogen during the heterogeneous radiolysis of water in the $RaO \cdot SiO_2 + H_2O_s$ and $RaO \cdot SiO_2 + H_2O_{liq}$ systems was studied. Two segments can be isolated: (i) on which the hydrogen accumulation rate on the initial linear segments is relatively high, (ii) with a relatively slow molecular hydrogen accumulation stage. The temperature effect on the rates of formation of molecular hydrogen during heterogeneous radiolysis of water was studied on the $(RaO)_x(SiO_2)_y + H_2O_s$ systems because in experiments on the radium-silicate+H_2O system in closed ampules the temperature cannot be increased [28]:

$$H_2O_s \xrightarrow[T \geq 373]{(RaO)x(SiO2)y} H_2 + 1/2 O_2.$$
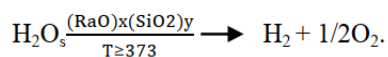
In the case of radium silicate and silica gel, all types of particles on the surface and in the bulk of water can be involved in water radiolysis. The generations of δ electrons that quit the solid phase possess sufficient energy for excitation of water molecules in the dissociative water level and initiation of radiolytic processes. Therefore, in heterogeneous radiation processes in the radium silicate +H_2O_{liq}, silica gel+H_2O_{liq} systems, the yield of molecular hydrogen is higher than in heterogeneous radiolysis of water in the absorbed states. The observed high yield of molecular hydrogen can be explaned by possible liberation

of nonequilibrium charge carriers G(n,p), excitons G(ex), and nonrelaxed and scattered electrons G (ege).

The radiation-chemical yield of hydrogen for radiumsilicate + H_2O (G(H_2] = 0.47 mol./(100 eV)) more than in the radiolysis of pure water (G(H_2) = 0.45 mol./(100 eV)).

The kinetics of accumulation of molecular hydrogen with radiation, the radiation-thermal and thermal (T = 300–673 K) processes radiumsilicate water [30].

It is shown that the formation of the surface-active centers and secondary electrons in the presence of radiumsilicate causes an increase in the saturation velocity of molecular hydrogen under thermal and radiation-thermal processes in the system radiumsilicate + H_2O [29].

The comparative analysis of Fourier IR - spectrums of absorption of nano-Zr+H_2Ovap and nano-Zr+H_2Oliq heterosystems shows that in both cases, radiation decomposition of water molecules is followed by the formation of an oxide film on the surface of zirconium.

In the first case it is possible to find and register absorption bands (AB) of intermediate and active products, an ion – radical groups π-O^{2-} and O_2^{2-}, and also surface hydrides.

In nano-Zr+H_2O_{vap} heterosystem, full decomposition of water molecules, and in nano-Zr+H_2O_{liq} system – partial decomposition of water occurs [28].

In work [30], the surface of zirconia particles was characterized by various spectroscopic techniques, and the variation in the surface following γ irradiation observed. Hydrogen production by γ-radiolysis of the mixture of mordenite, a zeolite mineral, and seawater was also studied by Yuta Kumagai et al. (2013) in order to provide basic points of view for the influences of zeolite minerals, of the salts in seawater, and rise in temperature on the hydrogen production by the radiolysis of water. These influences are required to be considered in the evaluation of the hydrogen production from residual water in the waste zeolite adsorbents generated in the Fukushima Dai-ichi Nuclear Power Station. As the mordenite's influence, additional production of hydrogen besides the hydrogen production by the radiolysis of water was observed. The additional hydrogen can be interpreted as the hydrogen production induced by the mordenite's absorbed energy at the yield of $2.3 \cdot 10^{-8}$ mol/J. The influence of the salts was observed as an increase in hydrogen production [31].

Based on these experimental results and further estimations, hydrogen generation dependent on decay heat and water content in the Submerged Demineralizer System vessel was finally revaluated. The procedure and results in this revaluation would help the decontamination of radioactive water at the Fukushima Daiichi NPP accident [32]. Studies performed at the VK-50 (operating a boiling reactor with natural circulation of the coolant) showed that the output of radiolytic gases with steam is largely determined by the power level and the quality of the coolant.

When analyzing the modes of the normal operation of the reactor, the coolant's radiolysis depends on the number of radioactive corrosion products in the coolant: [64]Cu, [65]Zn, and especially [59]Fe. With a pH change from 8 to 5,

the yield of radiolytic gases with steam increases from 60 to 200 nml/kg. In this case, radiolysis is significantly enhanced with a decrease in pH < 6 [33]. The relationships formed between the clusters suggest that the area of application of water radiolysis is expanding.

However, the overall management of radioactive waste is largely focused on the binding of liquid waste in matrix structures of different natures for long-term disposal (Fig. 8).
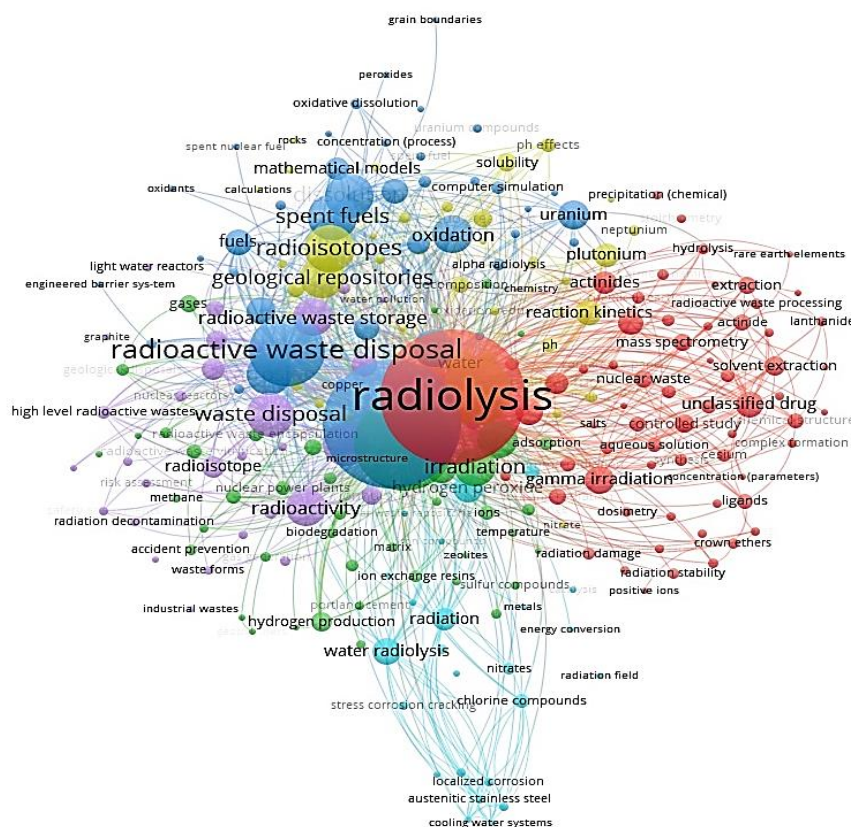

Figure 8 – General network visualization under application of radiolysis in the field of radioactive waste utilization (constructed via VOSViewer v.1.6.15): 5 clusters, 8344 links, 20038 total link strength

Figure 8 shows the clusters that were formed with using the VOSviewer: the red cluster includes research of radionuclide sorption processes and their concentration for radiolysis of liquid media; the green cluster concerns factors of influence on water radiolysis in hydrogen production; the yellow cluster includes research areas of natural processes related to radiolysis; the blue cluster is connected with mathematical modeling of radiolysis process with fuel production and engineering implementation and the purple cluster related to the processes of radioactive waste management and disposal as well as the application of radiolysis for this purpose.

Thus, research in the field of water radiolysis in connection with the problems of radioactive processing waste and the use of mineral systems with different sorption capacities to increase the yield of hydrogen is an urgent task nowadays. Also important is the technical implementation of the process, such as:

– designing methods to increase the uniformity of energy absorption of ionizing radiation;

– increase the penetration depth of low-energy electrons in the irradiated system;

– ensure turbulence of the irradiated volume of water, effectively saturate the liquid phase with gas;

– inject a gas-water jet into the volume of the radiation-chemical apparatus;

– regeneration volatile components of pollution;

– creating a closed circulation system in water treatment plants with electric accelerators thrones.

5 Conclusions

The ability to decontaminate the environment with an additional energy source is a leading environmental and energy challenge today. The situation with the processing and disposal of radioactive waste, including the liquid phase, is not resolved. Many questions remain regarding the environmental safety of processes and the technological implementation of the most energy-efficient solutions. Relationships in the field of research were visualized using VOSViewer. The development of new matrix materials combined with nanoparticles for the agglomeration and concentration of radionuclides is a promising innovation method improving radiolysis under hydrogen production from radioactive waste. The

development of new matrix materials combined with nanoparticles for the agglomeration and concentration of radionuclides is a promising method improving radiolysis under hydrogen production from radioactive waste.

This will allow additional study of new areas of liquid radioactive waste processing and their decontamination.

Further research will be focused on the following task is study of the directions of fixation of radionuclides and heavy metals in the matrix material of various genesis.

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