SCIENTOMETRIC STUDY ON HYDROGEN ABSORPTION ISSUE IN ZIRCONIUM ALLOYS USED IN NUCLEAR ENERGY

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Abstract

Zirconium (Zr) and its alloys have been used in various components of nuclear energy reactors for well over 50 years, during to the specific properties of the low neutron absorption section, good mechanical resistance under irradiation conditions and low corrosion at high temperatures. The main Zr alloys used in nuclear energy are Zircaloy-4 alloy (in fuel bundle element) and Zr-2.5%Nb alloy (in CANDU pressure tube). Hydrogen absorbed in these alloys, as a result of the corrosion reaction produced at the water-metal interface, can influence the thermo-mechanic behaviour of structural components due to the favouring of fragile platelets of Zr hydride. This, in turns, inevitably leads to the degradation of material properties under certain thermal and mechanical stress conditions. The process is influenced by the transport of hydrogen, the kinetics of precipitation and dissolution of Zr hydrides and the formation of hydrided microstructures specific to the texture of alloys. Precipitation of Zr hydrided platelets especially affects the ductility of the alloy and the tensile toughness, but it can also affect other phenomena by reorienting the hydrided platelets under certain thermo-mechanical conditions. The article aims to perform a scientometric study on the actuality of the hydrogen absorption problem in the reference alloys Zircaloy-4 and Zr-2.5%Nb, from the international journals and the Romanian literature, respectively, found on the Web of Science, Google Scholar, and Scopus platforms. A doctoral study is underway on this topic.

Keywords: *Scientometric study, Zirconium alloy, Hydrogen absorbtion, Zirconium hydrides*

Introduction

Zirconium alloys are used in nuclear reactors due to the combination of the following properties: high irradiation resistance, high corrosion resistance and low cross-section neutron absorption.

The present work is a scientometric study on the actuality of the hydrogen absorption problem in two Zr alloys used in nuclear energy (Zircaloy-4 and Zr-2.5%Nb) based on the international journals and the Romanian literature, respectively, found on the Web of Science, Google Scholar, and Scopus platforms [1 – 43].

The most demanding applications of these alloys in nuclear reactors are for nuclear fuel bundle tubes in PWRs (Pressurized Water Reactors), BWRs (Boiling Water Reactors), HWRs (Heavy Water Reactors) and PHWRs (Pressurized Heavy Water Reactors) as CANDU reactors, and for pressure tubes in RBMK (Russian acronym for Large-power channel type reactor) and other PHWRs. For the safe and economical operation of these reactors, both the fuel cladding and the pressure tubes should maintain their integrity throughout the lifetime of the reactor. However, during their residency in the reactor, these components are subject to ageing mechanisms resulting from thermal and pressure changes, rapid neutron bombardment and corrosion at the water/metal interface.

The latter results in a small part of the released hydrogen produced during the corrosion reaction being absorbed into the zirconium alloy. When the hydrogen concentration in the material exceeds the Zr-H solvent composition, Zr hydrides are formed. These Zr hydrides, which are less ductile than the surrounding metal matrix, may have harmful effects on the mechanical properties of Zr alloys, especially when present at a sufficiently large fraction of volume. The immediate effects are expressed by increasing the yield stress and decreasing the tensile strength of the Zr alloys.

There have been some early indications in the scientific literature that at relatively low hydrogen content in zirconium alloys, there may be a time-dependent cracking mechanism (now called Delayed Hydride cracking -DHC), as a consequence of a localized increase in hydrogen concentration in areas with high tensile stress values. This localized increase in hydrogen can lead to a crack and its subsequent propagation if there are favourable thermo-mechanical conditions.

However, even after the discovery that DHC mechanism caused leaks in some pressure tubes from two of the oldest operating CANDU reactors, the damaging mechanism was still not fully understood.

It was realized that this mechanism could pose a greater threat to the integrity of the pressure tube than the reduction of the overall tensile strength caused by the presence of zirconium hydride precipitation evenly distributed throughout the material.

As first scientific papers have shown, this embrittlement mechanism could only be important near the end of the life of pressure tubes, when there could be a sufficient increase in hydrogen in the alloy, so that Zr hydrides are present during normal reactor operating temperatures.

It has been found that the orientation of Zr hydride precipitation plays a much more important role in determining the ultimate tensile strength of these components than, for example, their volume fraction. This is because, at the hydrogen content of practical importance, Zr hydrides are formed as approximately linear rows of microscopic precipitate in the form of wafers on, or near the basal planes of the α - *Zr* matrix in which they are formed. In the fuel element sheath, Zy-4 alloy with high texture, and in the CANDU pressure tube, Zr-2.5%Nb alloy, these Zr hydrides predominantly have two tube orientations: circumferential and/or radial.

From the first investigations of Zirconium alloys used in fuel bundles and pressure tubes it was found that the fracture stress significantly deteriorated when the orientations of Zr hydride precipitation clusters were in the radial direction of the tube wall, compared to their orientations that were in the circumferential direction.

Methods for assessing the influence of hydrogen up-take on the Zirconium alloy material properties

The orientation of the hydrided platelets can have a strong effect on the ductility of the components made of zirconium alloys, namely Zircaloy-4 alloy used in the CANDU fuel element cladding and Zr-2.5%Nb alloy used in the manufacture of CANDU pressure tubes. For this reason, the ability to quantify hydrided morphology in terms of its effect on ductility is of essential importance in assessing the integrity of components made of zirconium alloys during normal operation in the reactor.

Several significant studies have been conducted over the past 60 years to better understand the formation and morphology of hydrides in Zr alloys. These studies show that the degree of degradation of ductility depends on the orientation, morphology and distribution of the hydride. It has been found that when hydride platelets are normally oriented to the direction of applied mechanical tension produce large reductions in the strength and ductility of Zr alloys, while if these are oriented parallel to the direction of applied mechanical tension have less effect (Fig. 1).

The experimental terminal solid solubility presents a hysteresis between the values obtained during heating and cooling (Fig. 2). The terminal solid solubility obtained during heating refers to the end of hydride dissolution and is called terminal solid solubility for dissolution (TSSD). The terminal solid solubility obtained during cooling refers to the beginning of hydride precipitation and is called terminal solid solubility for precipitation (TSSP).

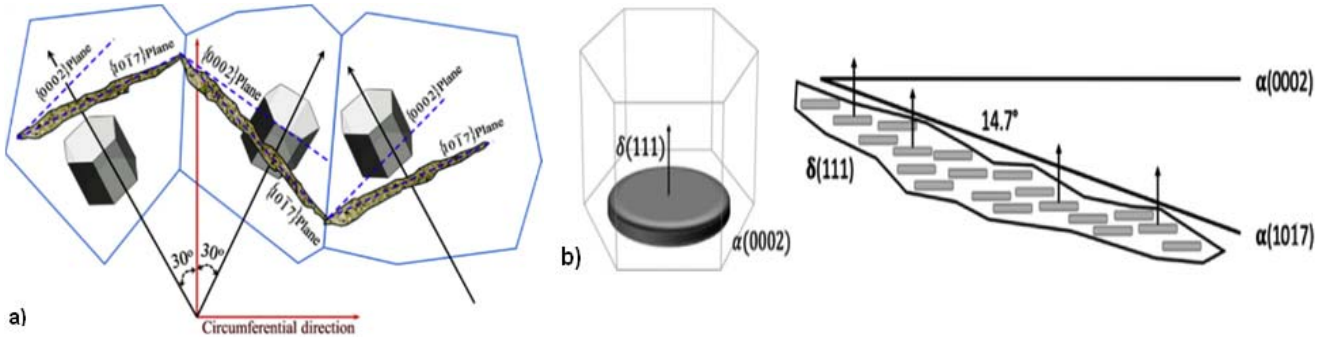


Fig. 1. The orientation of hydride platelets in the tubes made of Zr alloys (a), the structure of a macro-formed hydride and the orientation of the basal plane (b) [14]

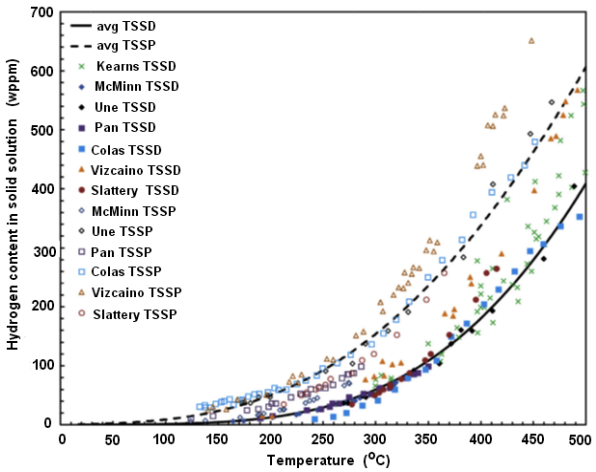


Fig. 2. Hysteresis on terminal solid solubility of hydrogen [44]

The nuclear fuel sheath made of Zy-4 is constantly cooled with water during reactor operation and spent fuel storage period. The reaction with water causes cladding oxidation and releases of hydrogen, which enters the tube wall and causes the Zr hydrides formation. During the reactor operation, nuclear fuel cladding is subject to constant circumferential stress caused by a combination of pressure created by fission gases produced inside the fuel and helium-filling gas from the fuel element. Other mechanical stress occurs during the transfer of spent nuclear fuel bundles to cooling storage, where temperature changes and internal pressure can cause re-precipitation and reorientation of the Zr hydride. In Fig. 3, phenomenon of re-orientation of the hydrides in Zircaloy-4 alloy cladding is presented under the circumferential mechanical loading.

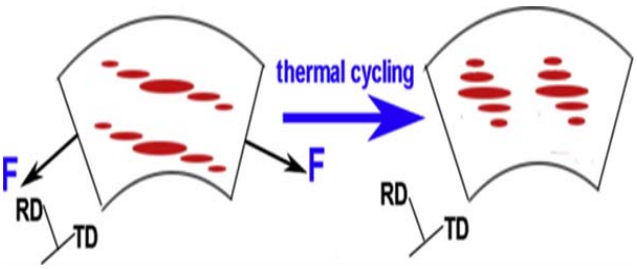


Fig. 3. The reorientation of the hydrides in Zy-4 alloy, [26]

The reorientation of Zr hydrides has a significant effect on the mechanical behaviour of the sheath and can cause its fracture and failure; this has been the subject of different experimental studies. The core of a PWHR (Pressurized Heavy Water Reactor) as CANDU (CANada Deuterium Uranium) consists of a network of horizontal pressure tubes which contains the fuel bundles. The pressure tubes from the CANDU reactors are periodically inspected according to the Canadian standard CSA N285.4. This standard specifies mandatory technical requirements and non-mandatory assessment procedures for fitness-for-service assessments.

During the reactor normal operation, the probability that a defect is generated on the interior inner surface of the CANDU pressure tube is quite large due to the following:
- the phenomenon of corrosion in contact with the heavy water flowing through the pressure tube;
- the vibrations induced by the flow of the cooling agent through the fuel bundles;
- the precipitation of hydrides through the continuous absorption of deuterium in the zirconium matrix;
- the generation of striations on the inner face of the pressure tube by the hard impurities trapped between the fuel bundle bearing pad and the pressure tube;
- the generation of blunt flaws by the fuel bundle bearing pad when the axial displacement of the fuel bundles occurs during the refuelling process.

When a defect is detected indicating that it does not meet the acceptance criteria by examination, or pressure tube to surrounding calandria tube contact is detected or predicted, Clause 12 of CSA N285.4 for the fitness-for-service assessment is applied for the next inspection interval. Also, Clause 12 mentions the need to assess the results of periodic inspections by the CSA standard N285.8.

There are three main characteristics of the microstructure of Zr alloys that contain hydride platelets and affect their structural integrity, namely:
- the total hydrogen content and the fraction of hydrogen that is precipitated in the hydride platelets;
- the fraction of hydride platelets oriented along the radial direction of the tubes;
- the continuity characteristic of the hydrides in the areas of interest in the thickness of the respective tube wall.

In the literature, several parameters have been defined to measure/characterize the morphology of Zr hydrides in the material volume (also called *metrics*) to describe its effect on crack initiation and propagation:
- Total hydrogen content;
- Volume fraction of zirconium hydride;
- Radial hydride fraction (RHF);
- Hydride continuity coefficient (HCC);
- Radial hydride continuity factor (RHCF).

The first metric referring to the Total hydrogen content can be experimentally measured. The second metric for the Zr hydride volume fraction can be estimated by using the metallography or X-ray diffraction. While these two metrics do not specify the hydride distribution, the other metrics refer more specifically to the morphology of the hydride in the material volume. The Radial hydride fraction (RHF) focuses only on the second characteristic; the Hydride continuity coefficient (HCC) and the Radial hydride continuity factor (RHCF) are used to evaluate the third characteristic.

The influence of hydrogen absorption as well as the formation of Zr hydride platelets in the Zr alloys used in nuclear energy, namely the Zircaloy-4 and the Zr-2.5%Nb alloys, can be realized by performing specific tests, which simulate the operation conditions for the structural components of interest (the fuel element sheath and the pressure tube). These tests can be grouped in the following study matrix:
1. **Tensile tests** to obtain mechanical properties and material constitutive equations, for example, the Ramberg – Osgood equation;
2. **Mechanical fatigue tests** to obtain the influence of hydrogen on lifetime, the Coffin - Manson equation;
3. **Mechanical overload tests**, to obtain the conditions for the initiation of cracking under mechanical tensile loading, the DHC phenomenon;
4. **Fracture mechanics tests** and obtaining fracture mechanics parameters represented by fracture toughness and $J - R$ resistance curve; Gurson-Needleman-Tvergaard modelling for prediction of crack initiation under mechanical stress;
5. **Microstructural analysis** (optical microscopy, scanning electron microscopy - SEM) and **Image Analysis Technique** to characterize the microstructural morphology and the reorientation of Zr hydrides.

In the literature, the hydrogen absorption effects in Zr alloys are a wide topic, which has reached a certain maturity, but still there are directions of study that can be addressed and improved.

Scientometric study of the issues related to Hydrogen absorption in Zr alloys

For the present scientometric study, after carrying out an extensive bibliographic study [1-43], the problem of hydrogen absorption in Zirconium alloys of interest in nuclear energy is focused on five directions of analysis:

- Zirconium alloys;
- Hydrogen absorption in Zr alloys;
- Precipitation of hydrides in Zr alloys;
- Reorientation of Zr hydrides in Zy-4 and Zr-2.5%Nb alloys,
- CANDU pressure tube.

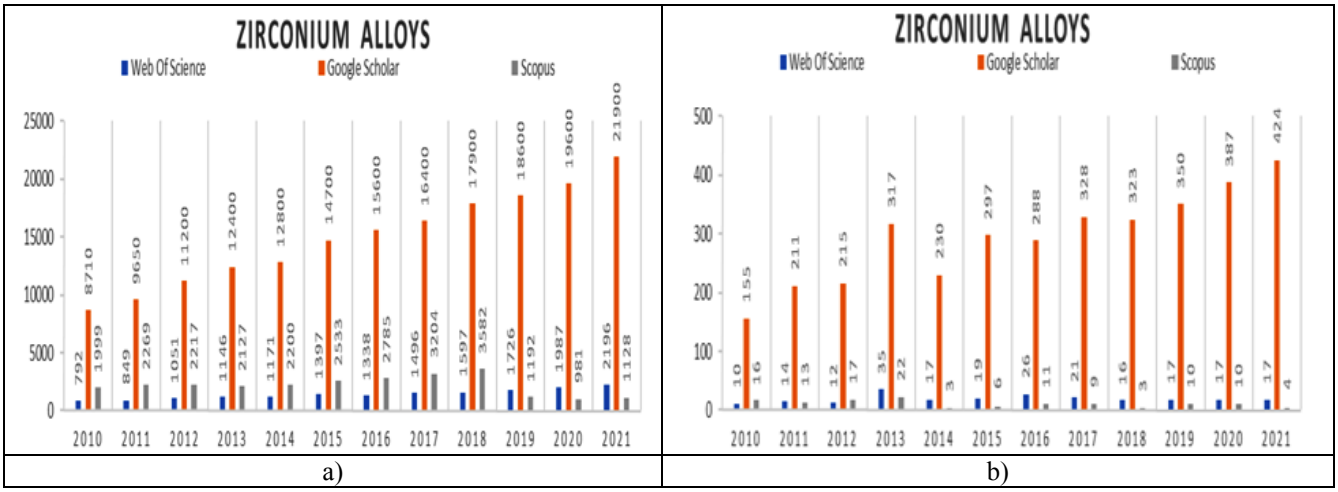
To carry-out the study, the scientific works published in specialized journals were counted in three database platforms, namely: Web of Science; Google Scholar; Scopus.

To pursue specifically the evolution of the number of studies dedicated to the directions of analysis, a period between 2010 and 2021 was chosen. Latter on, histograms representing the number of papers dedicated to above mentioned directions of analysis were drawn up. Meantime, the studies were divided into two categories, namely international studies and studies published in Romania.

The results of the study are given in Tables 1-5, the analysis histograms being represented in Figs. 4-8.

Table 1. Studies dedicated to the Zirconium alloys

Nr. crt.	Database	International	Romania	ZIRCONIUM ALLOYS											
				2010	2011	2012	2013	2014	2015	2016	2017	2018	2019	2020	2021
1	Web of Science	X		792	849	1051	1146	1171	1397	1338	1496	1597	1726	1987	2196
			X	10	14	12	35	17	19	26	21	16	17	17	17
2	Google Scholar	X		8710	9650	11200	12400	12800	14700	15600	16400	17900	18600	19600	21900
			X	155	211	215	317	230	297	288	328	323	350	387	424
3	Scopus	X		1999	2269	2217	2127	2200	2533	2785	3204	3582	1192	981	1128
			X	16	13	17	22	3	6	11	9	3	10	10	4



a) b)

Fig. 4. Histograms of the studies dedicated to Zirconium alloys: international (a), in Romania (b)

Table 2. Studies dedicated to the Hydrogen absorption in Zirconium alloys

Nr. crt.	Database	International	Romania	HYDROGEN ABSORPTION IN ZIRCONIUM ALLOYS											
				2010	2011	2012	2013	2014	2015	2016	2017	2018	2019	2020	2021
1	Web of Science	X		792	849	1051	1146	1171	1397	1338	1496	1597	1726	1987	2196
			X	3	6	8	14	6	5	8	9	5	10	8	7
2	Google Scholar	X		2660	3000	3600	4260	4080	4770	5280	6180	7050	8170	9580	12200
			X	155	211	215	317	230	297	288	328	323	350	387	424
3	Scopus	X		1999	2269	2217	2127	2200	2533	2785	3204	3582	1192	981	1128
			X	16	13	17	22	3	6	11	9	3	10	10	4

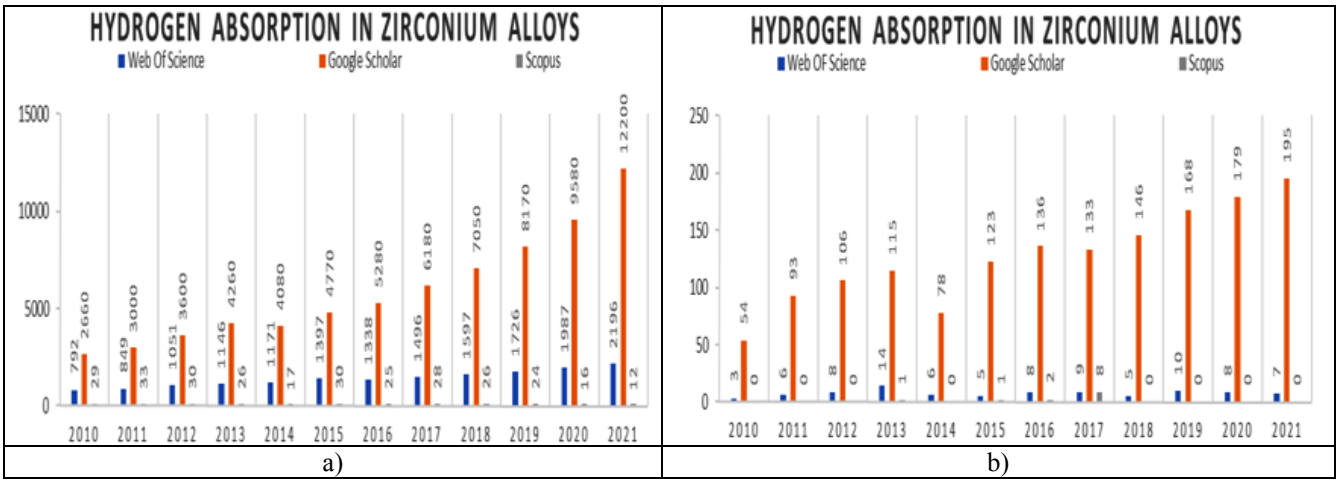


Fig. 5. Histograms of the studies dedicated to Hydrogen absorption in Zr alloys: international (a), in Romania (b)

Table 3. Studies dedicated to the hydride precipitation in Zirconium alloys

Nr. crt.	Database	International	Romania	HYDRIDE PRECIPITATION IN ZIRCONIUM ALLOYS											
				2010	2011	2012	2013	2014	2015	2016	2017	2018	2019	2020	2021
1	Web of Science	X		54	48	89	76	75	73	69	106	80	104	121	116
			X	1	3	5	8	3	4	3	1	0	3	1	1
2	Google Scholar	X		611	583	709	115	744	884	856	978	1050	1110	1310	1450
			X	13	24	34	33	13	23	22	23	22	20	28	35
3	Scopus	X		14	11	16	13	14	15	23	15	16	12	9	9
			X	0	0	0	0	0	0	0	0	0	0	0	0

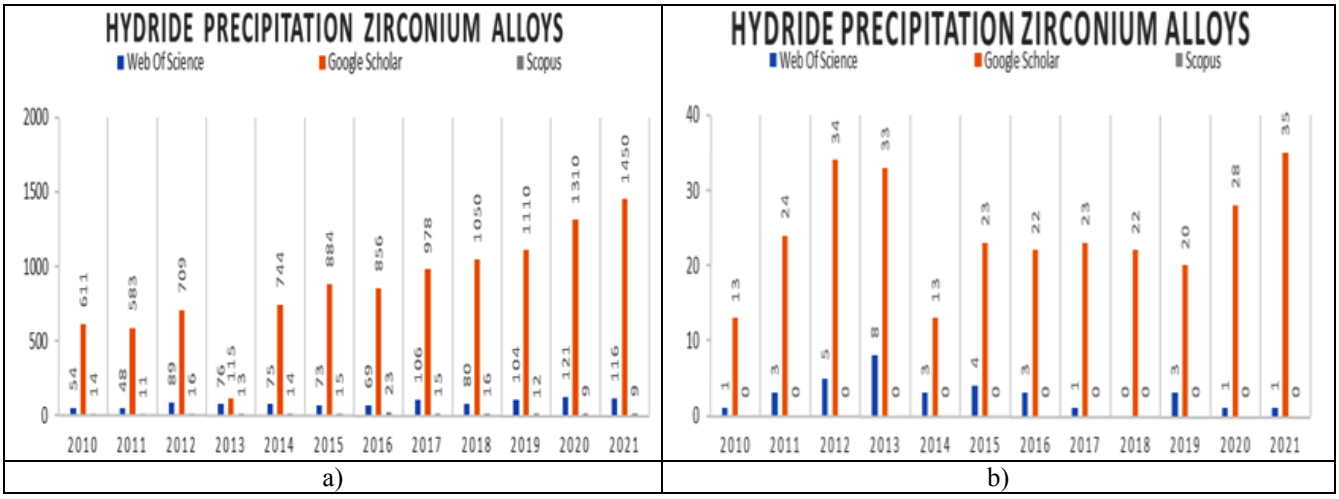
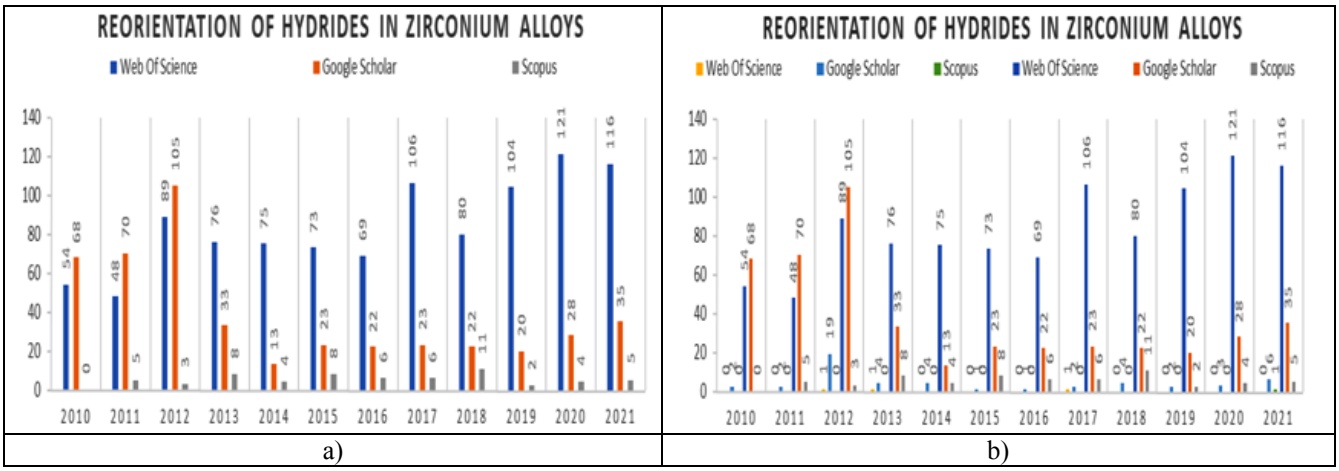


Fig. 6. Histograms of the studies dedicated to hydride precipitation in Zr alloys: international (a), in Romania (b)

Table 4. Studies dedicated to the reorientation of hydrides in Zirconium alloys

Nr. crt.	Database	International	Romania	REORIENTATION OF HYDRIDES IN ZIRCONIUM ALLOYS											
				2010	2011	2012	2013	2014	2015	2016	2017	2018	2019	2020	2021
1	Web of Science	X		54	48	89	76	75	73	69	106	80	104	121	116
			X	0	0	1	1	0	0	0	1	0	0	0	0
2	Google Scholar	X		68	70	105	33	13	23	22	23	22	20	28	35
			X	2	2	19	4	4	1	1	2	4	2	3	6
3	Scopus	X		0	5	3	8	4	8	6	6	11	2	4	5
			X	0	0	0	0	0	0	0	0	0	0	0	1

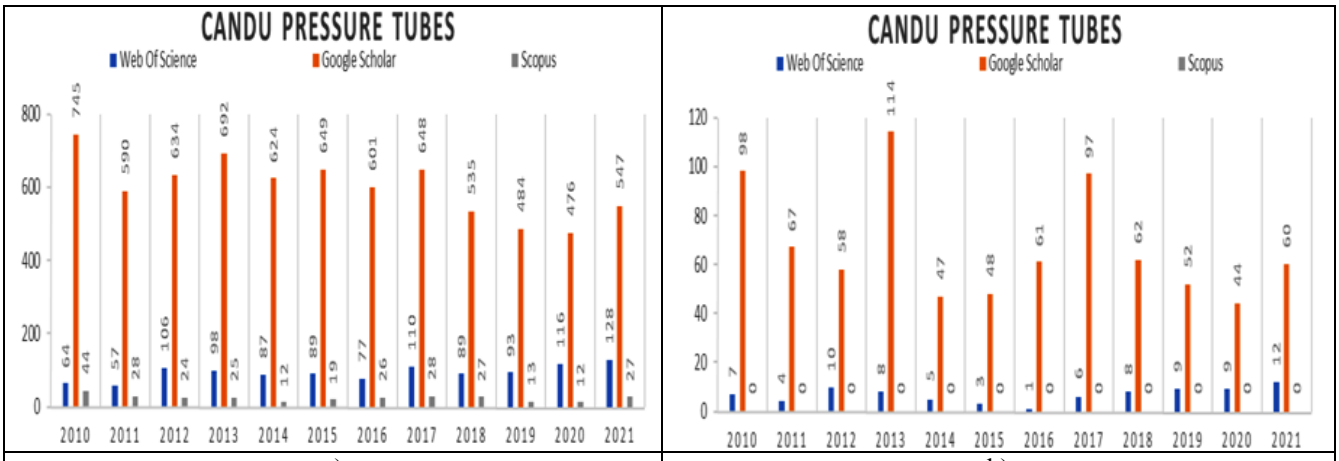


a) b)

Fig. 7. Histograms of the studies dedicated to reorientation of hydrides in Zr alloys: international (a), in Romania (b)

Table 5. Studies dedicated to the CANDU pressure tubes

Nr. crt.	Database	International	Romania	CANDU PRESSURE TUBES											
				2010	2011	2012	2013	2014	2015	2016	2017	2018	2019	2020	2021
1	Web of Science	X		64	57	106	98	87	89	77	110	89	93	116	128
			X	7	4	10	8	5	3	1	6	8	9	9	12
2	Google Scholar	X		745	590	634	692	624	649	601	648	535	484	476	547
			X	98	67	58	114	47	48	61	97	62	52	44	60
3	Scopus	X		44	28	24	25	12	19	26	28	27	13	12	27
			X	0	0	0	0	0	0	0	0	0	0	0	0



a) b)

Fig. 8. Histograms of the studies dedicated to CANDU pressure tubes: international (a), in Romania (b)

Some preliminary observations can be noticed here:
- the number of articles dedicated to each direction of analysis increases during the period selected for analysis as we approach of 2021, by about 2-3 times;
- the number of articles dedicated to the directions of analysis is high in the general case of the "Zirconium alloys" studies, and decreases with the specificity of the direction of analysis (from 1 to 5), the lowest number being registered for the reorientation of Zr hydrides and the CANDU pressure tubes directions, respectively;
- the number of articles published in Romania is much lower compared to the number of those published in international journals, and for certain directions of analysis it is even zero (the worst example is the reorientation of Zr hydrides, diection for which we have no published articles for several years in different databases);
- for Romania, which has two CANDU reactors in operation, one of them approaching the end of its designed life (Unit 1 from Cernavoda NPP soon will enter in the refurbishment), the subject is very topical.

Conclusions

After carrying out an extensive bibliographic screening on the problem of hydrogen absorption and its consequences in Zirconium alloys of energetic interest, a solid basis for the scientometric study was obtained. The embrittlement mechanism induced by the absorption of hydrogen, the formation of hydride platelets and their reorientation under the operating conditions of the structural components in the reactor, becomes increasingly important near the end of the reactor's life. When the reactor approaches the end of its operation life, there could be a sufficient increase of hydrogen in the alloy, so that zirconium hydrides are present during normal reactor operating temperatures.

The present work reviewed the problem of weakening Zr alloys through the formation of hydrides and established the coordinates of the scientometric study. Thus, the problem of hydrides hydrogen absorption in zirconium alloys of interest in nuclear energy is focused on 5 directions of analysis: zirconium alloys, hydrogen absorption in Zr alloys, precipitation of in Zr alloys, reorientation of zirconium hydrides in Zy-4 and Zr-2.5% Nb alloys, and CANDU pressure tube.

In this sense, the scientific works published in the profile journals were counted in three database platforms, namely: Web of Science, Google Scholar, and Scopus.

The scientific study follows the evolution of the number of studies dedicated to the proposed subject over a period of time between 2010 and 2021 for which histograms of the number of articles dedicated to the 5 directions of analysis were drawn up. At the same time, the studies were divided into two categories, namely international studies and studies published in Romania.

The analysis result revealed a number of articles published in Romania much lower compared to the number of international publications, and even zero for certain directions of interest (such as hydrides precipitation, reorientation of Zr hydrides, and the CANDU pressure tubes, respectively). That is why for Romania, which has two CANDU reactors in operation at Cernavoda NPP, with Unit 1 approaching the end of its designed life, the problem of hydrogen absorption and its consequences in Zr alloys of energy interest is actual and very important.

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