

На рис. 2 приведен пример диалогового отражения работы осциллографа с преобразователем вибрационных измерений. Из рис. 2 видно, что можно четко отследить амплитуду и периодичность изменения параметров вибрационного датчика, произвести запись вибраций и повысить точность исследования, используя масштабирование и изменение чувствительности.

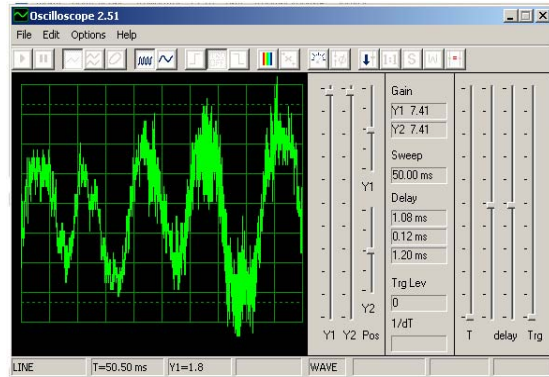


Рис. 2. Пример работы программы „Осциллоскоп 2,51” при подключении вибрационного датчика

**Выводы.** В результате проведенных исследований можно сделать следующие выводы.

1. Данная система анализа вибраций позволяет проводить исследование непосредственно на анализируемом узле.
2. При анализе использовать портативные компьютерные системы.
3. Есть возможность записывать вибрационные данные на внешние носители и в последствии анализировать на компьютерной технике.
4. Можно использовать независимое (автономное) питание.

Все вышеперечисленные возможности новой системы оценки вибрации подшипников не уступают современным приборам и устройствам того же направления и позволяют более тщательно провести анализ вибраций. По сравнению с приборами для анализа тихоходных подшипников данная система более мобильна и более дешева во время эксплуатации.

**Перспективы дальнейших исследований.** В дальнейшем вышеизложенным способом возможно проводить исследование не только технического состояния подшипниковых узлов, но и проводить исследования колебаний металлоконструкций и машин в целом. При использовании более совершенных вибродатчиков возможно также проводить исследования и диагностику работы зубчатых передач закрытого типа в различных механизмах. Также при небольшом преобразовании данным способом возможно снимать данные с тензометрических датчиков и исследовать динамику изменения напряжений в металлоконструкциях машин и механизмов.

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### AN INTEGRATED APPROACH TO THE ANALYSIS OF CREEP, CHEMICAL EXPANSION, CREEP DAMAGE AND LIFETIME REDUCTION FOR SOLID OXIDE FUEL CELLS

Теоретичні та чисельні дослідження даної роботи направлено на дослідження впливу явища електрохімічного переносу, дифузійної повзучості та хімічного розпухання в електрохімічній кераміці на ріст пошкоджуваності, що обумовлена повзучістю, та зниження часу безвідмовної роботи паливних елементів на твердому оксиді. Також в роботі досліджено, як керуючи електрохімічною дифузиею, процесом повзучості, хімічним розширенням та процесами пошкоджуваності в керамічних матеріалах знизити деградацію батарей нижче 0,1%/1000 годин, забезпечити оптимізацію паливних елементів, конструкції батарей та умов керування, збільшити термін роботи до 40000 годин при стаціонарному використанні та до 5000 годин при використанні в транспортних умовах.

In this paper, a comprehensive theoretical and numerical investigation has been carried out with the main focus directed at the understanding on how electrochemical transport phenomena, diffusional creep and chemical expansion in electrochemical ceramics affect creep damage growth and lifetime reduction for Solid Oxide Fuel Cell systems. Furthermore, focus is put on how the electrochemical diffusion, creep process, chemical expansivity as well as the creep damage growth in ceramic substances may be controlled in order to reduce stack degradation below 0.1% /1,000 hours, optimize cell, stack design and operating conditions, and extend stack lifetime above 40,000 hours for stationary applications and above 5,000 hours for transportation applications.

**Introduction.** The specific objectives of our international cooperation are:

- to identify the mechanisms of chemical, electrical and mechanical degradation that affect the lifetime reduction of Solid Oxide Fuel Cell (SOFC) systems, including ceramic cells, interconnects and stacks,
- to develop an integrated micro-meso-macro constitutive framework that will then be used to calculate the time dependent stress distribution and the creep

damage growth in SOFC stack under transient and steady state operating conditions as a function of material and stack parameters as well as operating conditions, and additionally to predict the lifetime of SOFC systems operating under severe service thermal and electrochemical conditions,

- to establish a relation between electrochemical transport phenomena, diffusional creep, chemical expansion and creep damage in electrochemical ceramics, and mechanical stress evolution in the stack and stack degradation over time,
- to incorporate an integrated micro-meso-macro constitutive model developed in our research into the ANSYS codes in a form of the computer-based structural modelling tool for analyzing mechanical stress distributions in the stack and stack degradation over time, for durability analysis and lifetime predictions, and for improving the performance and safety of SOFC systems for transport and stationary applications,
- to formulate practical recommendations based on the results of computational modeling and simulation for SOFC systems on how to modify materials, and improve stack design and operating conditions in order to reduce stack degradation below 0.1% /1,000 hours, and extend stack lifetime above 40,000 hours for stationary applications and above 5,000 hours for transportation applications.

**1. State of the art.** It is the fact that our world is facing the crisis of energy. The rapid depletion of natural resources and serious global environmental problems have become big concerns for the future. The decreasing fossil fuel supply has spurred an initiative to develop an alternative fuel to replace the carbon-based energy services. While some FCs require pure hydrogen, SOFCs can use many different fuels directly, including natural gas [1]. SOFCs developed at the end of the 20th century are composed of all-solid state materials that operate at temperatures around 1,000 degrees Celsius and push the efficiency of using fuel gas containing hydrogen to generate electric power to be around 50 percent of higher heating value [2]. SOFCs integrated in a gas turbine cycle can increase the total efficiency to 70 percent of higher heating value [2]. The electrochemical reaction in a high-temperature SOFC is carbon and oxygen (from ambient air) forming pure carbon dioxide and electric power.

SOFCs consist of an oxygen-ion conducting electrolyte material that is sandwiched in between two thin, porous electrodes. The cathode, anode and electrolyte are all made from ceramic substances. A schematic diagram of SOFC operation is given in Fig. 1 [3]. Different cell designs of SOFC have been studied [1], including planar, monolithic, segment-cell-in-series and tubular geometries. The cell component materials of the different designs are the same or similar in nature. It is vital that the different components (electrolyte, cathode, anode and interconnected material) have proper chemical and dimensional stability in oxidizing and/or reducing environments as well as chemical and thermal expansion compatibility with other components. Ytria-stabilized zirconia is the conventional electrolyte in SOFCs because of the adequate level of oxygen-ion-conduction at temperatures of around 1000 degrees Celsius and stability under both oxidizing and reducing conditions [1, 4, 5]. Nickel metal dispersed on the surface of a porous yttria-stabilized zirconia

support (nickel-doped yttria-stabilized zirconia) is used as the anode (fuel electrode) [1, 6], whereas porous lanthanum manganite doped with alkanite-earth elements is often used as the cathode (air electrode) [1, 7]. However, it was recently found [4, 7] that cathodes made of lanthanum ferrite provide two to three times more power at operating temperatures of 600 degrees Celsius to 800 degrees Celsius than those made of traditional cathode material, lanthanum manganite.

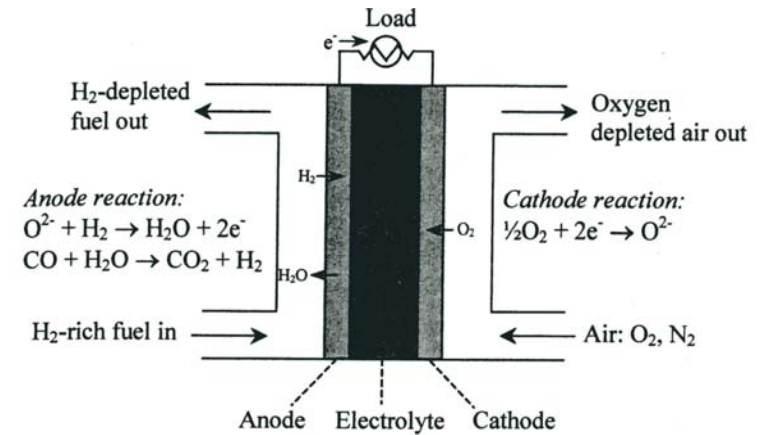


Fig.1. Operational principle of the power section of SOFCs [3]

Current research in the U.S.A and the European Union in the area of SOFC systems is aimed at reducing the cost per unit of electricity produced by an SOFC stack. For example, the long-term goal of the Solid State Energy Conversion Alliance (SECA) program by the U.S. Department of Energy is to develop low-cost SOFCs (\$400/kW) for stationary and transportation applications [2]. An important issue with respect to reduction of the cost per unit of electricity produced by an SOFC stack is the durability of the ceramic cell [8-10]. Therefore, it is necessary to gain a better understanding of the fundamentals behind the chemical, electrical and mechanical degradation that affect the lifetime reduction of SOFC systems, including ceramic cells, interconnects and stacks.

**2. Degradation of SOFC.** At the present, most of the FCs manufactures are concentrating on the anode-supported planar SOFC concept. However, a challenge with the planar geometry is in obtaining mechanically stable structure, as thin layer electrochemical ceramics are inherently susceptible to failure when subjected to thermal and mechanical stresses [11]. The magnitude of the stresses depends on the materials properties, the operating conditions and the geometry of the design. In 1999 at the Technology and Product Development Sector of Arthur D. Little Inc. for the cost assessment of anode-supported planar SOFC technology it was stated that the materials would survive any stresses arising from thermal gradients [12]. However, an SOFC manufacturing cost model was developed in 2004 within the SECA program that utilizes temperature gradients, and thermal and mechanical

stresses in SOFC stack to simulate relationships between performance, manufacturing and cost of production [12]. Actually, the stresses in SOFCs can arise from spatial or temporal temperature gradients, and from external mechanical loading. Stresses caused by the thermal gradients and by the difference between the thermal expansion coefficients and the Young's modulus of adjacent layers will increase with increasing cell area which conflicts with the desire to maximize the active cell area and therefore output. Thus, lifetime and durability of SOFC depend on the phenomenon on which the temperature has opposite influence: the right balance has to be found. In this context heat up and cool down are highly relevant since they can lead to very high thermal stresses. Furthermore, SOFC stacks are mechanically loaded with weights, during operation, in order to secure proper alignment and good contact between the cell components. This together with the seals required around the edges of cells to separate the fuel and air components, can cause higher mechanical stresses transmitted to brittle elements in SOFC stacks.

In the literature, there are a lot of papers, for example [13-15], that describe the general thermal-electrical planar SOFC performance. However, most of the authors do not consider the thermal stress modeling with exception of one [11, 16].

The consideration of the thermoelastic deformation of the SOFS system and the stresses resulting from temperature gradients and from external mechanical loading is quite important in the structural analysis of the system. However, this is not enough in order to understand the mechanisms of stack degradation over time that affect essentially the lifetime reduction of SOFC systems. Therefore, it is necessary to identify the time dependent phenomena related to the chemical, electrical and mechanical degradation of SOFC stacks over time. These time dependent phenomena can be investigated experimentally.

First, strontium-doped lanthanum ferrite  $\text{La}_{1-x}\text{Sr}_x\text{FeO}_{3-\delta}$  ( $\delta$  is oxygen non-stoichiometry), cathode material in SOFC under consideration, belongs to perovskite ceramics exhibiting both high ionic and electronic conductivity in which not only thermal expansion, but also another component, called chemical expansion, contributes to the overall observed expansion at high temperature [3, 17]. Thus, the chemical expansion observed in lanthanum ferrite at high temperature occurs in addition to the thermal expansion. This phenomenon can be explained taking into account that lanthanum ferrite must contain a high concentration of mobile oxygen point defects, and a decrease in the oxygen content of the perovskite will lead to expansion of the unit cell due to the larger size of the trivalent cation compared to the four-valent and weakening of the ionic bonds caused by removal of charged species [3]. Lowering the partial pressure of oxygen will also lead to increasing chemical expansion. Note that chemical expansion is not limited to perovskite materials, but can be seen as a general phenomenon concerning all mixed-valence materials that compensate for the reduction of cations by creating anion vacancies. In strontium-doped lanthanum ferrite, Sr substitution is necessary to obtain electronic and ionic conductivities of sufficient magnitude to meet the requirements to conduct oxygen ions. As the Sr substitution level increases, so does the potential for chemical expansion in oxygen potential gradient.

Figure 2 demonstrates the changing of the volume expansion of lanthanum ferrite of the cubic structure in air and nitrogen atmosphere with increasing temperature as dotted curves [3]. A broken line illustrates the expected volume expansion dependency on temperature in the absence of chemical expansion. The onset temperature of chemical expansion is defined as the temperature where the total volume expansion starts to deviate from linearity. Above the onset temperature the chemical expansion contribution to the total expansion increases with increasing temperature and strontium content. The extrapolations at high temperature are expected to coincide when the oxygen non-stoichiometry becomes equal in both atmospheres. At the temperature of 800 degrees Celsius in air chemical expansion is seen to account for approximately 33% of the total volume expansion of lanthanum ferrite. Decreasing the partial pressure of oxygen to 0.001atm increases the chemical swelling contribution to 50%. The same tendency for the chemical expansion changing over time has been found for different strontium-doped lanthanum ferrites [3, 18].

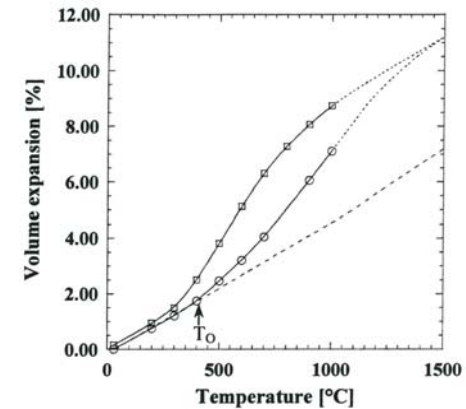


Fig.2. The volume expansion of perovskite  $\text{La}_{0.25}\text{Sr}_{0.75}\text{FeO}_{3-\delta}$  in air and nitrogen atmosphere

Under the operating conditions, SOFC is in a gradient of oxygen activity, and it is therefore subject to chemically induced straining across its thickness caused by the gradient in concentration of oxygen vacancies in the cathode material. This induces mechanical stresses that could ultimately cause fracture of SOFC. The chemically induced stresses depend on the variation of non-stoichiometry through the cathode under the operating conditions, the amount of chemical expansion for the given degree of non-stoichiometry and the geometry of SOFC [19]. Thus, chemical expansion of cathode material can cause the SOFC stack degradation over time and the lifetime reduction of SOFC systems. However, to the best of the authors' knowledge, up to now no investigations exist for chemically induced stresses distribution over time in SOFC stacks.

Second, another feature of electrochemical ceramics that can cause the stack

degradation over time and affect the lifetime reduction of SOFC systems is their creep behaviour. The creep deformation of yttria-stabilized zirconia (solid electrolyte) has been studied experimentally in [5, 20, 21] while the creep behaviour of nickel cermet anode has been investigated experimentally in [6, 9]. The creep deformation of strontium-doped lanthanum ferrite (cathode) has been studied experimentally in [3]. It is known [22] that creep deformation of electrochemical ceramics is related to the diffusion of the various species (cations and anions) in the crystal. Anion diffusion could be by oxygen vacancies or oxygen interstitials depending on the composition of the material. Grain boundaries are particularly likely to be affected by the presence of defects. The rates of mass transport processes, creep and grain growth can be controlled either by the effective diffusion coefficient of the slowest moving species or by the concentration of the slowest moving species. However, it is recognized that, in yttria-stabilized zirconia, oxygen anion diffusion occurs rapidly through the lattice and grain boundaries, and creep is controlled by the diffusion of the slower-moving Zr ion along either the grain boundaries or the lattice [20]. In strontium-doped lanthanum ferrite, the B-site diffusion, represented by Cr, was found to be faster than the A-site diffusion, represented by Y [18]. Thus, the consideration of diffusional creep of multi-layer (anode-electrolyte-cathode) is necessary to analyze SOFC stack degradation over time. Unfortunately, to the best of the authors' knowledge, in the literature there is only one publication [9] regarding the creep analysis of SOFCs. However, the electrolyte and cathode are assumed linear elastic without creep, and steady state creep in anode has been reached in about 200 hours. Thus, to the best of the authors' knowledge, up to now no realistic investigations exist for creep deformation of SOFC stacks. In 2005 it was stated that current research at Pacific Northwest National Laboratory within the SECA program is the beginning to include time-dependent behavior and properties for life-prediction studies of SOFC stacks [8].

Third, electrochemical ceramics in SOFC systems operating under severe service thermal and electrochemical conditions for a prolonged period of time exhibit creep deformation considered as a time-dependent irreversible deformation process. Even in the initial stages of the creep process in ceramics, dislocations, impurity atoms and voids accumulate at the grain boundaries to form grain boundary cavitation [3, 5, 6, 9, 20-22]. As microscopic cavities at the grain boundaries get larger and coalesce, dislocations, impurity atoms and voids move out to grain boundaries, and microcracks along the grain facets begin to be formed. Growth and coalescence of these microcracks lead to the creep rupture in the final stage of the creep process with formation of macrocrack with some preferential orientation, often, direction perpendicular to the maximum principal stress. Thus, creep deformation changes the microstructure of electrochemical ceramics by introducing dislocations, impurity atoms and voids in the initial stages, microscopic cavities in the following, and microcracks in the final stage of the creep process, all of them, at the grain boundaries with some preferential orientation. Furthermore, the velocity of the growth of already existing grain boundary microscopic cavities and microcracks, and of the nucleation of new ones essentially depends on the intensity of creep deformation.

On the other hand, creep deformation of electrochemical ceramics is influenced by the growth of microscopic cavities and microcracks. This influence begins at the primary and secondary stages of the creep process, and can be visible in the tertiary stage due to possible increasing of the creep strain rate, preceding the creep rupture. The creep rupture case without increase in the creep strain rate can be also observed in electrochemical ceramics. Thus, creep deformation and material deterioration in SOFC due to growth of creep damage occur parallel to each other, and they have a reciprocal effect. Obviously, creep damage growth in electrochemical ceramics leads to the chemical, electrical and mechanical degradation of SOFC stack over time. However, to the best of the authors' knowledge, up to now no investigations exist for creep damage development over time in SOFC stacks.

**3. New conception.** The interdisciplinary approach proposed in our research includes:

- the simultaneous consideration of electrochemical transport phenomena, thermoelastic deformation and chemical expansivity, as well as, diffusional creep process and creep damage development,
- constitutive modeling of these electrochemical and mechanical phenomena, and experimental determination of the tension/compression creep asymmetry for electrochemical ceramics,
- structural analysis of planar SOFC (anode made of 40 vol% Ni and 60 vol% 8YSZ, cathode made of Sr-doped lanthanum ferrite and solid electrolyte made from yttria-stabilized zirconia 8YSZ ) and SOFC systems using the ANSYS codes and constitutive model developed,
- computational modeling and simulation, and the quantitative analysis of stack degradation over time.

The planar SOFC stack of repeating unit cells with dimensions shown in Fig. 3 is under consideration. Total strains in SOFC are composed of an elastic part, thermal part, chemical part [23] and a part due to creep [24] related to a continuum damage parameter by Kachanov-Rabotnov [25] reflecting microstructural changes, material deterioration and stack degradation. Creep constitutive equation and damage evolution equation in our research take into account different properties of electrochemical ceramics under tension and compression. In order to investigate the effect of electric current on the chemical expansion of Sr-doped lanthanum ferrite, the simultaneous consideration of the Fick's second law and Nernst-Planck equation is given for the purpose of oxygen concentration determination. Analysis of stress distributions in the stack and stack degradation over time as well as life-prediction studies are related to the consideration of the physically nonlinear initial/three-dimensional boundary value problem. Therefore, various commercial software can be used for structural analysis, computational modeling and simulation, when the constitutive model proposed will be implemented into its codes. In accordance with recommendations given in [8, 13, 14], the ANSYS codes are accepted in our research. Experimental data for material properties necessary for determination of material parameters can be found in the literature [3, 5, 6, 8, 9, 13-15, 16, 18, 20, 21]. Creep experiments will be also conducted in the future in order

to determine the tension/compression creep asymmetry for electrochemical ceramics by residual stress measurements using laser technique, to find some material parameters needed in the constitutive model and to understand material behavior at the stack level.

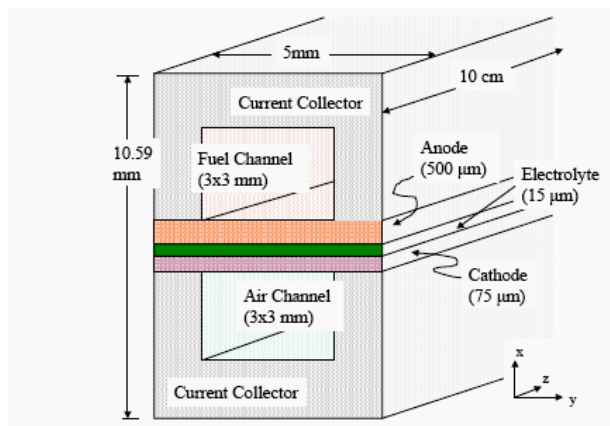


Fig.3. Geometry of the unit cell of a planar SOFC stack [15]

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