

Hydrogen Peroxide Decomposition as a Source of Propellant Gas for Actuators in Rehabilitation Robotics: a Feasibility Study

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Hydrogen peroxide decomposition is analysed for its possible implementation in a gas accumulation unit for the supply of a robotic actuation system. Reaction kinetics over MnO₂ powder catalyst were investigated and used for preliminary feasibility considerations. The results show that this process could be suitable for the considered application and its implementation is worth being further investigated.

1. Introduction

In the last decades, the field of wearable robotics has gone through a rapid growth, which has led to the development of devices that are today adopted for the rehabilitation and assistance of impaired subjects. In such applications, the lack of an autonomous and high performance actuation systems is an important issue that remains to be solved. Furthermore, from the intimate and continuous contact between machines and human body, some particular requirements in terms of lightness, safety and reliability arise.

The power supply systems and technologies that are currently used in this field include electromagnetic, hydraulic and pneumatic actuators. At present, these solutions are far from attaining the desired performances in terms of weight-to-volume ratio, power and energy density, bandwidth and controllability. With regards to pneumatic actuators, one major issue is bound to the need for heavy and bulky compressors or pressurized gas storage systems. One possible way to overcome this problem is to produce the pressurized gas by chemical reaction out of a liquid compound, which can be easily stored in a lightweight, small-volume vessel. The production of O₂ by catalytic decomposition of H₂O₂ shows good potentials for such application (Goldfarb et al., 2003; Vitale et al., 2010), mainly because of the safety of both reactants and products, high gas yields and reaction rates. The reaction, however, is strongly exothermic and the energy released during the decomposition may cause an excessive temperature increase that can be dangerous for the human operator. This issue becomes more relevant as the

propellant concentration in the reacting mixture increases. A possible solution is to use low-concentration H_2O_2 solutions, still keeping the required reaction kinetics by using appropriate catalysts.

In this work, a kinetic characterization of H_2O_2 decomposition catalysed by MnO_2 powder has been performed. To that end, experimental decomposition tests have been carried out in a constant volume, thermostated batch reactor.

The kinetic parameters of the H_2O_2 decomposition reaction have been used to assess the feasibility of a gas accumulation unit for the supply of an actuation system for assistance and rehabilitation devices.

The results obtained show that the H_2O_2 decomposition process, catalysed by MnO_2 , can be implemented in a gas storage and production unit complying with the size and weight constraints imposed by the specific application scenario. Therefore, the H_2O_2 decomposition process represents a viable alternative to standard power supply systems in this field and is worth to be further investigated.

2. Materials and Methods

Hydrogen peroxide solution 30% w/w was purchased from Sigma Aldrich (Italy). Manganese dioxide powder was purchased from Carlo Erba Reagenti (Italy) and used as catalyst.

Experimental runs of H_2O_2 decomposition were carried out in a stainless steel closed reactor, provided with a liquid inlet and a gas outlet nozzle, both equipped with a stop valve. The reactor had an available internal volume $V_{tot} = 320.6$ ml and was equipped with pressure and temperature sensors connected to a data acquisition system. In order to control the temperature, during the experimental tests, the reactor was immersed in a thermostatic bath; furthermore, water from the thermostatic bath was also circulated in a coil located inside the reactor.

In each experimental run, the reactor was initially loaded with a given mass of MnO_2 powder and then sealed and immersed in the thermostatic bath. At the beginning of each experiment, both valves were opened, about 100 ml of H_2O_2 solution of known concentration were injected in the reactor and, immediately after, both valves were closed. Temperature and pressure time courses were recorded during each run.

The experiments were carried out with different temperatures of the thermostatic bath (25°C and 50°C), catalyst mass (3.5 and 7.8 mg) and initial H_2O_2 concentrations (1.17, 1.73 and 3.46 M). A summary of the experimental trials carried out is presented in Tab.1. Due to the strong exothermicity of the decomposition reaction, the temperature in the reactor and thermostatic bath could differ slightly, especially in the early stages of the tests; the actual temperature time course measured inside the reactor was taken into account in data analysis.

Table 1: Summary of experimental runs (experiments marked with * were not used for parameter estimation)

Data set	T [°C]	M [mg]	C_0 [M]
Mn035-25-12	25	3.5	1.14
Mn035-50-12	50	3.5	1.10
Mn078-25-18	25	7.8	1.70
Mn078-50-18	50	7.8	1.68
Mn078-25-36*	25	7.8	3.46
Mn078-50-12*	50	7.8	1.10

3. Mathematical Model

Data collected in the experimental trials were analysed by means of a mathematical model that was derived under the following main hypotheses: 1) both phases in the reactor behave as ideal mixtures; 2) gas absorption in the liquid phase is negligible; 3) H_2O_2 evaporation is negligible; 4) in each instant, H_2O is in vapor-liquid equilibrium conditions. The model equations include the mass balances for the three components in the reactor, the ideal gas equation of state and the vapor-liquid equilibrium condition for H_2O :

$$-\frac{dn_p^L}{dt} = \frac{dn_w^L}{dt} + \frac{dn_o^G}{dt} = 2\frac{dn_o^G}{dt} = k_0 \exp\left(-\frac{E_a}{RT}\right) M \frac{n_p^L}{v_p n_p^L + v_w n_w^L} \quad (1)$$

$$P[V_{tot} - (v_p n_p^L + v_w n_w^L)] = (n_o^G + n_w^G)RT \quad (2)$$

$$\frac{n_w^G}{n_w^G + n_o^G} P = \frac{n_w^L}{n_w^L + n_p^L} P_w^s(T) \quad (3)$$

In eq. (1)-(3) P is the pressure, T the temperature, P^s the vapor pressure, n the number of moles, v the molar volume; subscripts w , p , o refer to H_2O , H_2O_2 and O_2 , respectively; superscripts G and L refer to the gas and liquid phase, respectively. Furthermore, M is the catalyst mass, k_0 and E_a the Arrhenius pre-exponential factor and activation energy of the decomposition reaction, respectively.

Initial conditions for the differential variables in eq. (1)-(3) can be easily determined by knowing the initial pressure in the reactor, volume and composition of the liquid phase. Since the temperature time course was measured experimentally is known, an energy balance was not included among the model equations and the algebro-differential system (1)-(3) can be numerically integrated to calculate $n(t)$ for each component in the two phases and $P(t)$.

Equations (1)-(3) contain only two unknown constants, namely k_0 and E_a , that were used as adjustable parameters to fit experimental data.

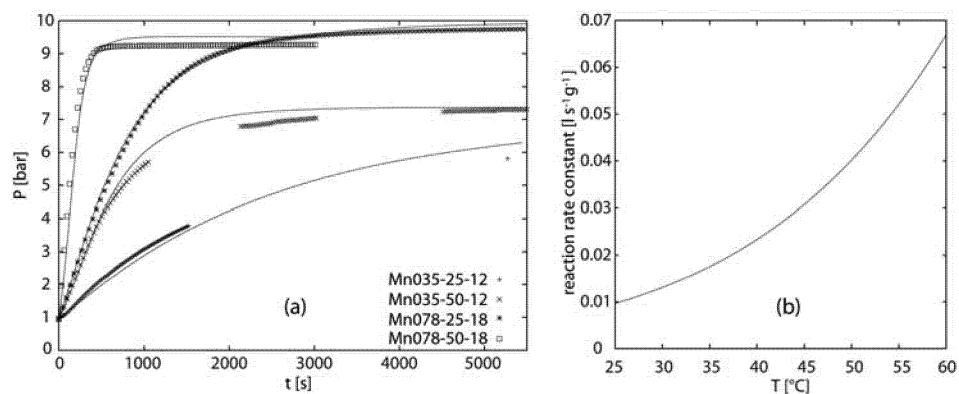


Figure 1.: (a) Pressure time courses: experimental (points) and calculated with optimal parameter determined by model fitting (lines). (b) Optimal temperature dependence of the reaction rate constant.

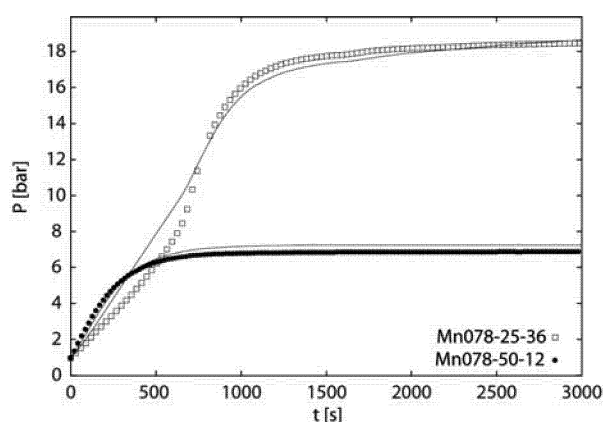


Figure 2.: Model prediction (lines) compared to experimental data (points) of two experiments

4. Results

Pressure time courses recorded during the experimental runs are reported as points in Fig.1. In each experiment, the pressure increases with time because of the production of gaseous O_2 , until the H_2O_2 initially loaded in the reactor is completely decomposed. Clearly, the higher the initial H_2O_2 concentration in the solution, the higher the final pressure in the vessel.

The model presented in section 3 was fitted simultaneously to the pressure data of four out of the six experiments carried out (see Tab.1) by using k_0 and E_a as adjustable

parameters. Figure 1b, shows the optimal temperature dependence of the reaction rate constant obtained by the fitting procedure.

Calculated pressure time courses obtained with the optimal parameters are reported as lines in Fig. 1a. It may be seen that the agreement between the experimental data and the calculated curves is satisfactory.

As a further verification of the results obtained, in Fig. 2, data from experiments that were excluded from parameter estimation are compared to the prediction of the model, again showing good agreement.

5. Feasibility of a Gas Accumulation Unit Based on MnO₂ Catalysed H₂O₂ Decomposition

The characterization of the kinetics of H₂O₂ decomposition reaction can be used for a preliminary analysis of the implementation of this process in a gas accumulation unit for the supply of robotic actuators.

Even if in this work the actual design of such accumulation unit is not considered, the preliminary calculations that will be performed need the definition of a reference configuration of the device. Such a device may consist of a small volume vessel, i.e. 1.5 l, with an available internal volume of 0.9 l. As a tradeoff between propellant accumulation and pressure buffering capabilities, 67% of the available volume can be considered filled with aqueous H₂O₂ solution.

The accumulation unit must comply with the instantaneous needs of the actuation system, supplying gas at constant pressure while the flow-rate may continuously change from zero up to its maximum value. To that end, the unit may be equipped with a control system able to throttle the gas production rate. One possible way of throttling the H₂O₂ decomposition rate is by changing the amount of catalyst immersed in the liquid solution, for instance by lowering or raising a catalyst bar inside the liquid.

As for the process specifications of the accumulation unit, an operating pressure of 4 bar seems suitable for actuation needs, also accounting for pressure drops in the distribution system, while gas flow rate depends on the actuation system coupled with the accumulation unit and working conditions. A set of three FESTO DG 2456 alternative single effect actuators (Festo Corporation, Hauppauge, NY, USA), each with a piston displacement of 0.71 ml, working at a constant frequency of 2 Hz will be considered here. In these operating conditions, that can be considered as demanding for the actuation system, the gas flow rate needed by the actuation system is 4.6 ml/s, measured at 4 bar and 25°C, corresponding to an O₂ production rate (neglecting, for the sake of simplicity, H₂O and H₂O₂ vaporization) $F=0.69$ mmol/s. In order to keep the reaction rate constant while H₂O₂ is consumed, the immersed mass of catalyst must be increased progressively. The needed mass can be estimated by integrating the H₂O₂ balance in the liquid phase (1), with the simplifying assumption of constant volume of liquid V_L :

$$M(t) = \frac{2F}{k_0 \exp(-E_a/RT)(C_0 - 2F/V_L t)} \quad (4)$$

where C_0 is the initial concentration of H₂O₂. Clearly, the needed catalyst mass tends to infinity as time approaches a value t^* when H₂O₂ is exhausted. Assuming $C_0=3.5$ M,

which is close to the concentration of some over-the-counter H₂O₂ solutions, $t^* \approx 0.5$ h and, after 25 min, $M \approx 0.7$ g. These figures are more than reasonable considering that the actual operating conditions would be milder and higher concentrations H₂O₂ could be used.

These results show that H₂O₂ decomposition process represents a viable alternative to standard power supply systems in the field of wearable robotics and is worth to be further investigated.

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