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I. NUCLEAR REACTORS FOR II HYDROGEN AS A FUEL: SOME ASPECTS HYDROGEN PRODUCTION. OF ITS COMBUSTION PROCESSES.

REACTEURS NUCLEAIRES POUR LA PRODUCTION D'HYDROGENE.

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L'HYDROGENE COMME COMBUSTIBLE:

OUELOUES ASPECTS DES PROCESSUS

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PART I: NUCLEAR REACTORS FOR HYDROGEN PRODUCTION

1.1. INTRODUCTION

The oil crisis has been considered a warning sign, calling attention to the problem of the limited availability of our energy resources, specially in view of the rapidly increasing use of fossil fuels in the next past years. It is hoped that \gg the future the increase in oil demand - which was 7,2% annually during the past years -will decrease in relation to the following factors:

- a) progressive utilization of other energy vectors.
- b) finalization of the process of substituting coal by oil*
- c) introduction of measures directed towards a more rational utilization of energy.

However, the assured world oil resources wiI I only cover the foreseen needs until the decade of 1990. After that the introduction of improvements in oil extraction procedures and large capital investments will be required with the result of an important increase in cost.

As far as natural gas is concerned, and although the United States and Western Europe covered mainly their requirements with domestic resources, there are sufficient indications to believe that the situation has started to change and that soon the economic conditions regarding this product will be similarto those referring to oil today.

In view of the considerably high coal reserves in many industrialized countries, sometimes the opinion is held that this fuel can cover, on a medium-term basis, a great part of the energy demand in the western world. However, due to the physical properties of coal and the steps which will have to be taken for environmental protection before it could be used, pre vious treatment and transformation to other more suitable synthetic fuels will be necessary.

In addition, transformation of coal into liquid or gaseous fuels requires large quantities of energy, if its infrautilization is to be avoided.

Therefore, new alternate sources of energy which subs_ titute fossil fuels will be required. Modern technology is undoubtedly inclined towards nuclear energy as a substitute for fossil fuel. In the beginning and for several reasons, nuclear energy was developed almost exclusively for obtaining electricity. However, we must not forget the difficulties involved in transmission, distribution and specially in storing electricity as a form of energy.

A look at the total energy consumption in its different facets shows that the electricity represents a minor fraction in the whole concept, with heating and transportation representing the greatest percentage. For this reason, the application of nuclear energy for obtaining process heat for later application, such as obtaining hydrogen, is already foreseen. These medium-term forecasts do not contradict the present plans for the immediate futurs, wherein the use of nuclear energy for electric energy production will be intensified.

With regard to nuclear fuel, we wish to point out that the world resources *are* important and *are* widely distributed. The assured resources at a price of less than \$ 30 a pound of U.Oo the world's foreseen needs up to 1990.We should in addition remember the small incidence of the cost of nuclear fuel in the price of the kWH generated which will permit economb utilization of deposits considered in principle as non-profitable. On the other hand, the introduction of breeder reactors will multiply, by a considerable factor, the available uranium resources. Also, the thorium cycle which uses this element as a fertile material in nuclear reactors presents itself-as very promising.

The combined use of nuclear energy as a primary source of energy and of hydrogen as *art* energy vector, because of its transport and storage characteristics, seems to offer ideal prospects which will make possible the substitution or replacement in the consumption of a great part of fossil fuels.

1.2. NUCLEAR ENERGY APPLICATION IN HYDROGEN PRODUCTION

1.2.1. General hydrogen production methods.

As hydrogen is not found in a free state in nature, it must be obtained from those materials which contain same in their composition, such as: coal, hydrocarbons and water.

Hydrogen production procedures can be divided into two categories: a) open-cycle processes and b) closed-cycle processes. In the open-cycle processes, hydrogen is extracted using fossil carbon or hydrocarbons and heat. The foMowing are obtained as reaction products: hydrogen, CO^{*} and degraded heat. In the closed-cycle processes, water and neat are used and they produce: hydrogen, oxygen and degraded heat. This second type corresponds to the electrolytic and thermochemicaI processes for obtaining hydrogen* We are outlining below, in general terms, the most important procedures for obtaining hydrogen and the peculiarities of using in said processes the energy produced in nuclear reactors.

1.2.2. Open-cycle processes.

Steam reforming of natural gas is based on the following endothermic reaction:

^{CH}4 ^{+ H}2° ~~~ ^{C0 + 3H}2

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Since the heavier hydrocarbons are present only in small proportion.

The CO shift reaction also takes place:

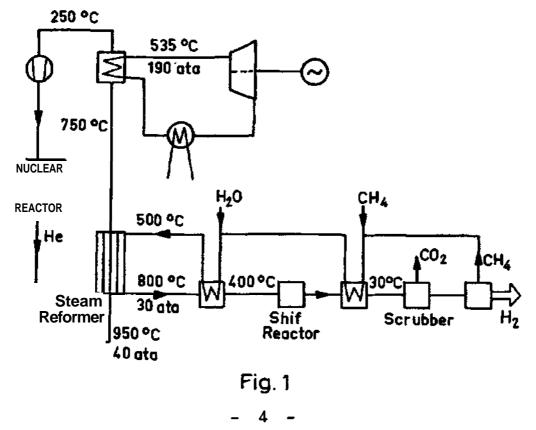
 $CO + H_2O$ $CO_2 + H_2$

As a consequence, the net reaction is as follows:

 $Ch + 2H_{/} 0 - * C0_{,,} + 4H_{,,}$

The above is carried out in a vertical tube furnace, which contains a catalyst (for example nickel). The heat is applied externally and the gases at the outlet of the reformer have a temperature of 7002 to 9002C. The procedure is we I I-known and the technology is developed on an industrial level. At the present time, the majority of the hydrogen produced in Europe and the United States is produced with this procedure. The heat supply is obtained by burning coal.

Methane conversion with steam in the reforming plant can also be obtained by supplying heat coming from helium gas at 9503C, which is used as a coolant in a High Temperature Gas Reactor, HTGR, (see figure 1)»



The HTGR are thermic reactors moderated with graphite and cooled with helium, and excellent coolant as it does not absorb neutrons and it is more inert than C0". The sire of the HTGR reactor is considerably smaller than the British advanced gas reactor (AGR), basically due to the high degree of fuel enrichment, which results in a decrease in capital costs. The fuel is formed by small particles of uranium and thorium dicar_ bide, coated with three layers of pyrolitically-deposited graphite and one layer of silicon carbide to prevent the release of fission products. The particles are cast into rods using a carbonaceous binder in order to obtain the fuel pellets which are inserted in prismatic graphite clusters, in which openings are made for fuel elements, control rods and for coolart; passage. The final arrangement of the blocks is in a cylindrical lattice which gives shape to the reactor's core. The refractory property of the core and the matrix of the fuel is the main characteristic which permits operating at high temperatures without producing an important contamination in the primary circuit with fission products.

The charecteristics of a dual purpose HTGR reactors (production of hydrogen and electricity) are identical to the commercial HGTR reactors which only produce electricity. How ever, the most significant differences between the dual purpose and single application HGTR reactors are the following: a) higher helium temperatures at the core's outlet (95Q2C as compared to 760-C in the electricity-production reactors). These temperatures of between 900⁹ and 1000SC are achieved without having to increase the maximum fuel temperature which is approximately 1250°C, b) incorporation of helium-heated steam-methane reformer in a cavity made in the reactor's vessel wall, c) inclusion of ducts for helium circulation between the reformer and the steam generator. The output temperature in the reformer is approximately 7502C and the pressure 25 atmospheres. In this field the German pilot plant EVA (Einzelrohr-Versuchs-Anlage) could be mentioned. This plant operates under similar conditions to those which could be obtained using a HTGR, The helium is heated by a electrical procedure which simulates the heat generated in a reactor core.

The second procedure for obtaining hydrogen -in the category of open-cycle processes - in other words, coal

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gasification, is presently being developed and is based on the following reaction:

 $C + H_2 0 - CO + H_2$

which together with the shift reaction:

$$CO + H_2O_{--} - CO_2 + H_2$$

gives the following net reaction:

 $C + 2H_20 - \gg \bullet CO_2 + 2H_2$

This procedure presents fewer environmental problems than the conventional gasification processes. Gasification takes place at temperatures between 650[^] and 850[^]C. The energy required for the steam-coal reaction can be obtained starting from nuclear process heat. The range of temperatures needed is more suitable to the present HTGR technology than those required for methane reforming.

1.2.3- Closed-cycle processes.

As the availability of fossil fuels will decrease with the passing of time and their cost will progressively increase, it is expected that, on a long-term basis and once hydrogen has been introduced on the market, it should be obtained from other materials such as water. For these reasons, present technology is directed towards establishing or improvising procedures, starting only from water, for obtaining hydrogen, such as: electrolysis and water decomposition by means of thermochemicaI processes.

The use in such process of electric power generated in a nuclear reactor does not mean any special problem. Research work in this field is basically directed towards improving the electrolysis process specially as for its thermal efficiency, with the development of appropriate electro Iyzers and electrodes.

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Included in the closed-cycle processes, in addition to electrolysis, the procedure for obtaining hydrogen -thermo- ' chemically offers the greatest interest for future introduction in the hydrogen production industry. This is based on economic considerations of process efficiency and capital costs In a thermochemical process, all the elements and compounds which take part in the different reactions are regenerated, with the final reaction being:

 $H_20 - K_2 + 1/2 0_2$

The water decomposition cannot be carried out by means of simple thermolysis, as partial pressures of hydrogen and oxygen, in equilibrium with water steam, at the atmospheric pressure and at a temperature of 2000°C, are very low. However, dissociation is possible by means of indirect methods.

As the thermochemical process is a high-temperature chemistry problem, it must be examined from this point of view in order to understand its theoretical limitations in that whicf refers to efficiencies for the different reaction temperatures and for identifying the ideal values in enthalpy and entropy changes.

The practical thermochemical cycles require more than 2 reactions or stages. The decomposition temperatures, for the cycles recently proposed, are located in temperature ranges between 6002 and 1000^{fi}C. At present an important research work is being carried out in different centers and laboratories of Europe, U.S.A. and Japan, many of them related with nuclear research.

Present research is directed towards determination of thermodynamic data at high temperatures and of kinetic parameters which, in their majority, are unknown. From the technological point of view, development work contemplates compatibility and material strength, as the products which take part in the reactions- are usually very corrosive or hard to handle. It also attempts to establish chemical separation techniques and suitable heat exchange systems. The objectives are: to select the most simple process possible in order to reduce the heat required; decrease the investment cost of the chemical plant and achieve the greatest possible thermal efficiency.

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The more appropriate nuclear reactors to supply the heat required for this process are again HTGR. Nevertheless, on safety reasons, it does not seem advisable, at least at the present time, the direct flow of the nuclear reactor coolant through the process heat exchanger, thus making it convenient to use an intermediate heat exchange loop. The introduction of. this loop will no doubt result in the need to raise the fuel temperature in the reactor core. Nuclear technology needs some improvements in that which refers; to the fuel and core design, as temperatures required for the process will be much higher, about 1300SC.

The application of nuclear power in the production of hydrogen through thermochemical processes presents very pro mising prospects, although adequate development work is required both in our knowledge of the thermochemical processes themselves as well as in that referring to the nuclear reactors for produv cing the necessary heat. Regarding the latter, technology corres_ ponding to a new high temperature reactor design, up to about 13009C, is required. This would basically mean from a nuclear point of view a new design of fuel element, and from a conventional technological point of view - high temperature heat exchangers suitable to the process.

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PART II

HVVROGEH AS A FUEL; SOME ASPECTS OF ITS COMBUSTION PROCESSES

II. 1 INTRODUCTION

The possibility of utilizing hydrogen as a fuel in air breathing power plants or in other energy conversion systems in a near future has been extensively studied and discussed, as for example, in the First World Hydrogen Energy Conference, including the associated problems of energy storage and energy transportation.

Research on combustion of hydrogen has been mainly pionered through aerospace programs, due to the fact of the high interest of hydrogen both as a propellant for liquid fueled rocket motors, and as a fuel of excellent characteristics for air breathing engines, for the propulsion of subsonic, supersonic and hypersonic vehicles.

Interest in hydrogen has increased considerably due to the energy crisis, and consequently research programs on its combustion, production, transportation, liquefaction, etc. as well as on its utilization in conjunction with nuclear and solar energy have increased enormously in the last few years.

At the Instituto Hacional de TScnica Aeroespacial "Es_ teban Terradas" (INTA) of Spain, a modest but continuous research programs on hydrogen combustion-have been carried out for several years, which are still in progress.

A large part of this research effort has been sponsored by the Air Force Office of Scientific Research of the United States and another part of the programs by the "Fundaci6n Juan March" of Spain, In the present work some results of these research programs are shown, specially those related to some specific properties of the hydrogen, which differ considerably from those of typical hydrocarbons fuels (Table II.1).

Hydrogen is an excellent fuel for both jet and reciprocating engines. Its characteristics of high combustion temperature, high calorific value, very low flamroability limits, smokeless flame and very good cooling properties are well known and have been widely discussed elsewhere, in connection with engine life, reduction of energy consumption, emission of pollutants, utilization in supersonic airplanes, etc.etc.

Other properties of the hydrogen such as high values of its transport coefficienus, resulting in high diffusivity, very low value of its critical pressure and practical abscense of free convection in combustion processes taking place within hydrogen atmospheres originate several very characteristics problems which have been studied at INTA, and will be discussed in the following paragraphs.

	'I'ABLE II-I			
	Boiling Temp. (1 atm). °K	Critical Temp. °K	Critical Pressure, atm.	
Hydrogen	20	33	12.8	
Methane	112	190	45.8	
n-Octane	399	370	t2.0	
Air	77 (N ₂ >	126	33.t	
	88 (0 ₂ >	155	50.0	
	Thermal con- ductivity cal/sec ² cm x	Viscosity coef. centipois	Diffusion coef. in air cm^/sec.	
	x °K/cm.			
Hydrogen	0.170	0.085	0.611	
Methane	0.020	0.0105	0.390	
n-Octane	0.097	0.550	0.0505	
Air	0.016	0.0185	_	

TABLE II-l

II.2 COMBUSTION OF LIQUTV PKOPELLANTS IN A HVVROGEH ATMOSPHERE

One of the first research programmes on hydrogen conbustion carried out at INTA has been the theoretical and experimental studies on combustion of liquid oxidizers in droplet form within a hydrogen atmosphere. This is the basic process controlling- combustion of hydrogen-oxidizer sprays, since due the fact of the low critical pressure of hydrogen and of the proximity of its boiling and critical temperatures, hydrogen is usually injected as a homogeneous fluid into the combustion chambers, that is to say, no hydrogen droplets exist during the combustion process.

The theoretical program, as shown in refs.l through 6 comprised the study of the flame structure and temperature, droplet burning rate, and flame radius. Model of the process considered quiasi-stationary conditions, spherical symmetry and both finite and infinite chemical kinetics reaction rates. The equation of the process were solved by means integral methods.

Numerical applications were carried out for the cases

of bromine, oxygen and nitric acid droplets. Some typical re suits are shown in Figs. II-1 y II-2 (refs. 2, 3 and 6).

The experimental program comprised the measurements of the droplet burning rates and flame radius by means of normal and high speed cinematographic cameras of bromine and nitric acid droplets within hydrogen atmospheres at pressures ranging from i to 100 K/cm².

Due to the low density of hydrogen free convection ef fects were almost non-existant, and therefore, spherical flames were obtained, as shown in the photographs of Fig.II-3, and some typical experimental results are shown in Fig.II-»».

Bromine droplets exhibited an explosive type of combus tion, as shown in the high speed photographs of Fig.II-5.

A part of the theoretical program was devoted to the study of the influence on the process of the values taken for the transport coefficients, which depend considerably on the composition in all hydrogen-oxidizer combustion mixtures. This effect complicate considerably most theoretical studies.

Some of the principal results obtained are summarized in Table II-2.

An extension of this work was carried out by studyng hydrogen-air flames by using porous spheres through which hydrogen was injected and burned in air or conversely. The theoretical treatment is shown in refs. 4 and 5 and some experimental results are given in Figs. II-6 and 2-7.

II-3 <u>COMBUSTION OF HVVROGEN IN SUPERSONIC AIR STREAMS</u>

During the period 1960-1970 a research program was carried out at IMTA on supersonic combustion problems with particular emphasis on the hydrogen combustion. The reason for this interest has to be found on the hypersonic vehicle concept denominated SCRAHJET (Supersonic Combustion Ramjet). Spain worked on the subject through Grants with the European Office of Aerospace Research of the United States Air Force.

Due to the nature of this vehicle, for which high com bustion temperatures and rapid mixing and burning of fuel in the air stream are required, hydrogen is the most suitable fuel.

Among all the proposed systems of supersonic combustion, the purely diffusive mode seemed to be the one that fits better the large flight Mach numbers (6-25) and the mod_ el usually employed in these works consisted of a low velocity hydrogen jet injected downstream into a faster coflowing stream.

	IADIE II-Z		
	Dimension- less burning rate x _s "\» m/r _s	<pre>Evaporation constant k= -dr /dt cm²/sec,</pre>	Flames dro- plet radius ratio r*/rs
Oxygen droplets in hydrogen (constant transport coeffi- cients)	14.00	3.27×10^{-3}	2.10
Oxygen droplets in hydrogen (variable transport coeffi- cients)	2.80	5.79×10^{-3}	1.78
Nitric acid droplet* in hydrogen (theo- retical results)	1.02	«•.«* x 10 ⁻³	1.53
Nitric acid droplets in hydrogen (exper- imental values)		6.5×10^{-3}	1.30
Bromine droplets in hydrogen (theoreti- cal)*	13.2	12.6 x10 ⁻³	1.095

TABLE II-2

Detailed calculations of the simultaneous laminar mix ing and combustion processes were carried out in refs. 7 and 8, that were extended to the turbulent case in ref.9. In ref. 10 a more refined model of the laminar one is presented.

Simple analytical methods for the analysis of the chemical kinetic effects in turbulent diffusive combustion were given in ref.ll, showing the existence of three different regions (fig.II-8). The first region is close to the injector and the flow may be considered frozen. Far from the injector the flow is in chemical equilibrium and in between there is a transition region. Integral methods for treating the equili-

* No experimental values of burning rates and flame radius were available due to the explosive type of combustion. Comparison with experimental results were made through ignition regions a function of pressure as shown in ref.l. brium region are given in ref.12.

In refs. 13 and 14 the ignition stage is studied. The ignition delay is calculated considering a constant pressure, free mixing layer, where fuel and oxidizer mix without appreciable concentration change resulting from chemical reactions. The field near the injector is further complicated by the recirculation due to adverse pressure gradients induced by the wake-like configuration (fig.H-9) that appears at low ignition rates and by the aerodynamic effects of the heat released from the flame, once it has been established. This recirculation improves the mixing process.

II-4 NITKOGEN OKIVES FORMATION

The only pollutants emitted in hydrogen-air flames are nitrogen oxides, especially under the form of NO. Therefore, it is very important to study the formation of such components in $H_{\mu}-O_2-N_2$ flames.

A study of this kind was carried out at INTA sponsored by the "Fundacio'n Juan March", and some of the results obtained were presented at the 9^{tn} World Energy Conference (ref.16).

Starting from a kinetic scheme for the H2-O2-N2 reaction composed of 34 elementary reactions, the princial results of the research program was the derivation of over-all reaction rates of the NO, at temperatures around 1000°K and 3000°K, and at pressures from 3 to 40 atm., which may be utilized for the calculation of NO production in adiabatic combustion processes under given conditions.

II-5 SUPERCRITICAL COMBUSTION

Supercritical combustion of fuels and propellants in droplet form has become a problem of high technical interest since the operating pressures in the combustion chambers of rocket motors and jet engines have reached values higher than the critical pressure of several fuels or oxidizers, such as kerosene, oxygen and especially hydrogen, which has an unusually low critical pressure (Table II-1).

As discussed in ref. 17, fuels and propellants may be injected into the combustion chambers at supercritical pressure but at subcritical temperatures. Surface tension is a function of droplet temperature becoming equal to zero as tem perature reaches its critical value. Therefore, droplets for mation will follow injection and the initial combustion or vaporization process may take place at supercritical pressure and subcritical temperature. Therefore, droplet temperature will increase till it reaches its critical value, and then both surface tension and the latent heat of vaporization will become equal *to* zero, and the droplet will become a sphere of fluid surrounded by a mixture of the same fluid and combustion gases.

On the oth-e-r hand, hydrogen is/normally injected at pressures and temperatures higher than both critical values, since it is usually utilized as a coolant prior injection. Therefore the supercritical combustion process of hydrogen with oxygen (or other oxydizer) will take place as a spray of oxygen fluid spheres within an homogeneous hydrogen atmosphere.

The process of supercritical combustion has been studied at INTA, and the results published in refs. 17, 18 and 19. Equations of the process were solved both numerically and by means of an asymptotic analysis. In Figs. 11-10 and II-ll some of the results obtained are given. Fig. 11-10 shows dimensionless extinction times as function of the temperature far from the droplet (T°) , initial droplet temperature (T_{Q}) , and hydrogen concentration at large distance from the droplet (Y_{206}) . Finally, Fig. 11-11 shows a numerical application for the H2-02 case, giving combustion times of oxygen droplets of 10^{-2} cm initial diameter for the complete subcritical and supercritical fields. It shows that combustion time is minimum, precisely at critical pressure, theoretical prediction which has been experimentally verified.

II-6 ACTUAL MSEAKCH PROGRAMS

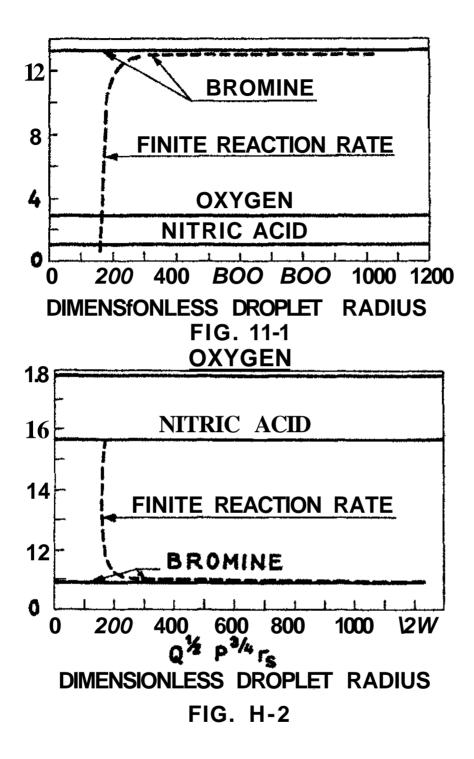
Present research programs on hydrogen combustion are being continued in two different fields;

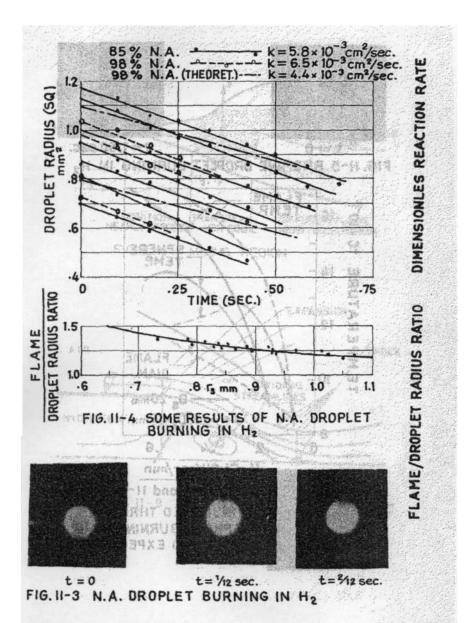
Forced convection and non-stationary effects on super critical combustion of droplets, with an application to the hydrogen case, are being studied under a Grant of the U.S. Air Force of Scientific Research.

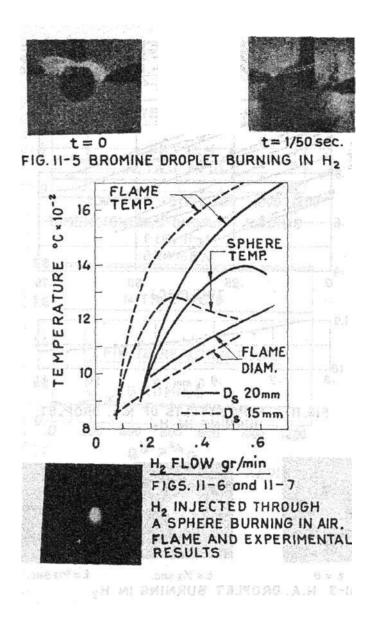
In collaboration with the "Escuela TScnica Superior de Ingenieros AeronSuticos" of the "Universidad Politecnica" in Madrid, a research program is being conducted oir" the problem of injecting small amounts of hydrogen into the combustion chamber of a one-cylinder variable compression ratio reciprocatins -engine, with the aim of utilizing leaner mixtures in order to reduce both emission of propellants and energy consumption.

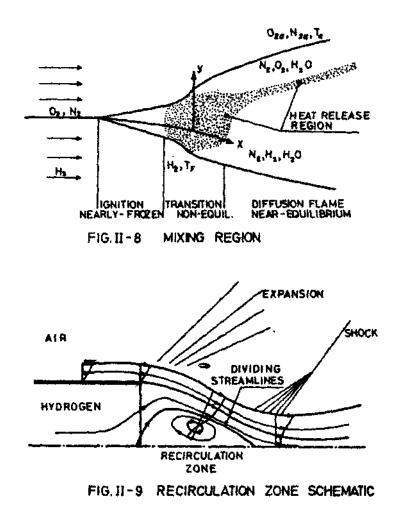
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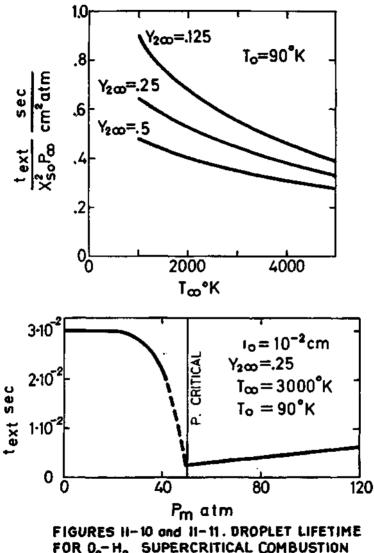
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FOR 0,-H, SUPERCRITICAL COMBUSTION AND COMPARISON WITH SUBCRITICAL CASE

ABSTRACT

The present work is divided into two parts: In the first part a brief summary is presented of the applications of nuclear energy to hydrogen production. In the second one some aspects of the combustion processes of hydrogen as a fuel are presented and discussed.

One interesting application contemplated for nuclear reactors is hydrogen production. The most important procedures for obtaining hydrogen and the peculiarities of using in said processes the energy produced in nuclear neac tors with the present state of this technology are reviewed.

Research programs carried out in Spain on combustion of hydrogen are reviewed. Results of combustion of several liquid oxidizers in droplets within hydrogen atmospheres and the study of the resulting spherico-symmetrical flames *are* discussed, as well as the flames resulting from the injection of hydrogen in air through porous spheres.

The problem of mixing and combustion of hydrogen in supersonic air streams are shown, in connection with the modelling of supersonic combustion ram-jet vehicles.

Results of nitrogen oxides formation in H_2 -air flames are shown in connection with pollution problems.

Supercritical combustion of oxygen in hydrogen is discussed showing results and including an outline of the present research program.

Finally, the problem of injecting small amounts of hydrogen in spark-ignition reciprocating engines is shown in conection with the aim of reducing both energy consumption and emission of pollutants which is one of the present research programs.

RESUME

Ce rapport est divise en deux parties. La premiere contient un bref resume des applications de l'energie nucieaire a la production d'hydrogene. Dans la deuxifeme que Iques aspects des processus de combustion de l'hydrogene en tant que combustible sont presented.

La production d'hydrogene est une des applications interessantes prevues pour les reacteurs nucteaires. On fait un expose des principaux processus pour la production d'hydrogene et des particularit£s d'employer dans cea processus l'6nergie produite dans les reacteurs nucleaires, ainsi qu'une r6fe>ence sur I'etat actuel de cette technologic

Les programmes de recherche qui se realtsent en Espagne sur la combustion de l'hydrogene son signales. On discute les resultats de la combustion de differents oxidants Iiquides en petites goutes dans une atmosphere d'hydrogene et l'etude des flammes de symetrie spherique qui se sont produites ainsi que les flammes provoquees par l'injection d'hydrogene dans l'air a travers sphferes poreuses.

Les problemes du melange et de la combustion d'hydrogene dans des courants d'air supersoniques en connexion avec ('adoption de modeles pour stato-reacteurs supersoniques de combustion, sont presented.

Les resultats de la formation d'oxides de nitrogfene dans des flammes H«-air par rapport aux problemes de contamination sont signales.

On discute la combustion supercritique d'oxygene dans un milieux d'hydrogene exposant les r6sul"tats obtenus. Le programme de recherche actuel est presente dans ses Iignes generales.

Finatement on fait reference au problfeme d'injecter de petites quantit6s d'hydrogene dans des moteurs alternants d'allumage electrique afin de reduire tant la consommation d'energie que l'6mision de contaminants, &tant devenus a present I'objet d'un des programmes de recherche.