Operating Experiences of Mercury Collection by PAC Injection in Bag Filters.

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ABSTRACT

Efficient control of mercury emission from waste incineration flue gases is well-established technology and it is the result of a history where different approaches were tried out. At first sight, this application appears different from that prevailing at coal-fired boilers, but in some plant configurations the chemical conditions are similar, and the technical problem is similar to that of coal-fired boilers. In Europe, several incinerators have been equipped with fabric filters of the ALSTOM Filsorption type with PAC injection downstream scrubbers and reheat where the HCl has been reduced to some few ppm and mercury has been reduced back to its elemental state by the upstream scrubber sulfite oxidation. These flue gas conditions are similar to those downstream an ESP on a coal-fired boiler. In this work, the operating results from the waste industry are compared to results of short-term tests of PAC injection on coal-fired boiler flue gas under similar conditions. From this experience, the coal-fired boiler industry can benefit from several years of both performance data as well as practical operational experience.

INTRODUCTION

The problem of mercury control at coal-fired power boilers is a central topic of the present conference, and several technologies - in various stages of maturity - are being discussed.

Injection of PAC (powdered activated carbon) in existing ESP's has been tested in full scale in some projects [1, 2] and the result was typically that 60-85 % mercury collection efficiency was obtained at a PAC flow of 15 lbs PAC/Macf. To get a perspective, this figure corresponds to a fly ash carbon content on a PC boiler corresponding to a LOI of 3-4 %, or an approximate PAC cost of $1.5/MWh$_e$ ($0.5/lb PAC).

Injection of PAC into a fabric filter with a very low ash influx at the Gaston Station (the fabric filter being located downstream an ESP having 97-99 % fly-ash collection efficiency) , [3], gave 90% mercury collection efficiency at a PAC feed rate of 3 lbs/Macf corresponding to a PAC cost of about $0.3/MWh_e$. In such an arrangement the ESP ash is not affected and different reutilization schemes are conserved.

It appears that the factor limiting the performance of PAC-injection upstream an ESP is the diffusive mass-transfer from the gas phase to the PAC- particles in suspension, [4] . This gas absorption case is singular in the power industry in that the mass of sorbent is low (order of 50-200 mg/Nm^3). Correspondingly, the mean distance between the sorbent particles is large (the order of 2mm for 20 microns PAC particles), and the residence time for the sorbent in the gas before the sorbent is separated from the gas in practical plants is short (2 secs.) . In a bag filter cake, the gas residence time is short (0.2 sec.) but the distance between gas and sorbent particles corresponds to the cake pore channel size which is of the order of some 20 microns. Denoting the diffusion distance x, and the contact time t, we find that the quantity x/sqrt(t), (which is the scaling parameter in the one-dimensional diffusion equation), we find that for the two cases these quantities are:

Injection upstream the ESP, $x/\sqrt{t} = 1\ mm/sec^{1/2}$.
In a filter cake $x/\sqrt{t} = 0.05\ mm/sec^{1/2}$

This illustrates how the diffusion-limited mass transfer process of mercury to PAC particles proceeds much further in a fabric filter case than in the ESP case.

In a wet limestone/gypsum desulfurization systems, the mercury chloride in the gas is absorbed and dissolved in the massive flow of scrubbing liquid:

$$HgCl_2 (g) \rightarrow HgCl_2 (aq.) \rightarrow Hg^{++} + 2Cl^-$$

However, the massive flux of absorbed SO_2 (10^5-10^6 times the flux of mercury) gives a corresponding production of sulfite which is partly oxidized already during the residence of the drop in the flue gas:

$$SO_3^{--} + OH^- \rightarrow SO_4^{--} + H_2O + 2e^-,\ E_0 = +0.90\ V, \text{ or}$$
$$H_2SO_3 + H_2O \rightarrow SO_4^{--} + 4H^+ + 2e^-,\ E_0 = -0.17\ V$$

The oxidation of sulfite can drive the reduction of mercury ions to metallic mercury:

$$Hg^{++} + 2e^- \rightarrow Hg^0,\ E_0 = +0.85\ V,$$

since the sum reaction in both cases gives a positive emf. figure.

This then leads to release of metallic mercury vapor to the flue gas:

$$Hg^0 (aq) \rightarrow Hg^0 (g),$$

It is indeed observed that the emission of mercury from WFGD systems does consist of metallic mercury vapor even when a large fraction of the mercury in the flue gas prevails in the oxidized state on the system inlet side, [5].

It then appears there is an inherent conflict between on one side one of the very basic chemical processes in a limestone/gypsum WFGD system and on the other side the system's potential to control mercury emissions.

By exclusion, one can then conclude that the most promising technology for efficient mercury control on power boilers is the fabric filter. The PAC injection function is proven in full-scale tests, but not yet in continuous operation. From a power industry perspective, it could then be of interest to scrutinize the experience of PAC injection into fabric filters in other industries.

THE WtE EXPERIENCE

In the 80's the waste-to-energy industry in Europe faced radically increased pressure to reduce CO, NO_x, particulates, HCl, HF, SO_2, dioxins and mercury. Different suppliers took different approaches to solve this technical task. The mercury problem consisted of controlling a concentration in the 500-800 $\mu g/Nm^3$ from the boiler to an emission level of 10-50 $\mu g/Nm^3$. At least the following techniques were explored in full scale:

- acid scrubbers
- acid and oxidizing scrubbers
- sulfide and tetrasulfide system
- zeolite adsorption
- coke beds
- PAC injection into fabric filters
- and combinations of the above

In the natural selection process of the real-life application world, costs, reliability and performance has made the market to converge to acid scrubbers to reach the higher emission levels (50 $\mu g/Nm^3$) and additionally, PAC injection into fabric filters to reach the lower emissions levels (10 $\mu g/Nm^{3)}$.

Consequently, PAC injection in dedicated fabric filters for mercury control is well-established technology for WtE plants in Europe. The flue gas composition and plant sizes admittedly differ from the conditions on coal-fired boilers, but the technical problem also has many features in common to the problem of mercury control on coal-fired boilers. There is also an experience basis of a decade-long successful continuous operation on more than a dozen plants. The flue gas cleaning system in these larger incinerators in Europe is shown in Fig 1.

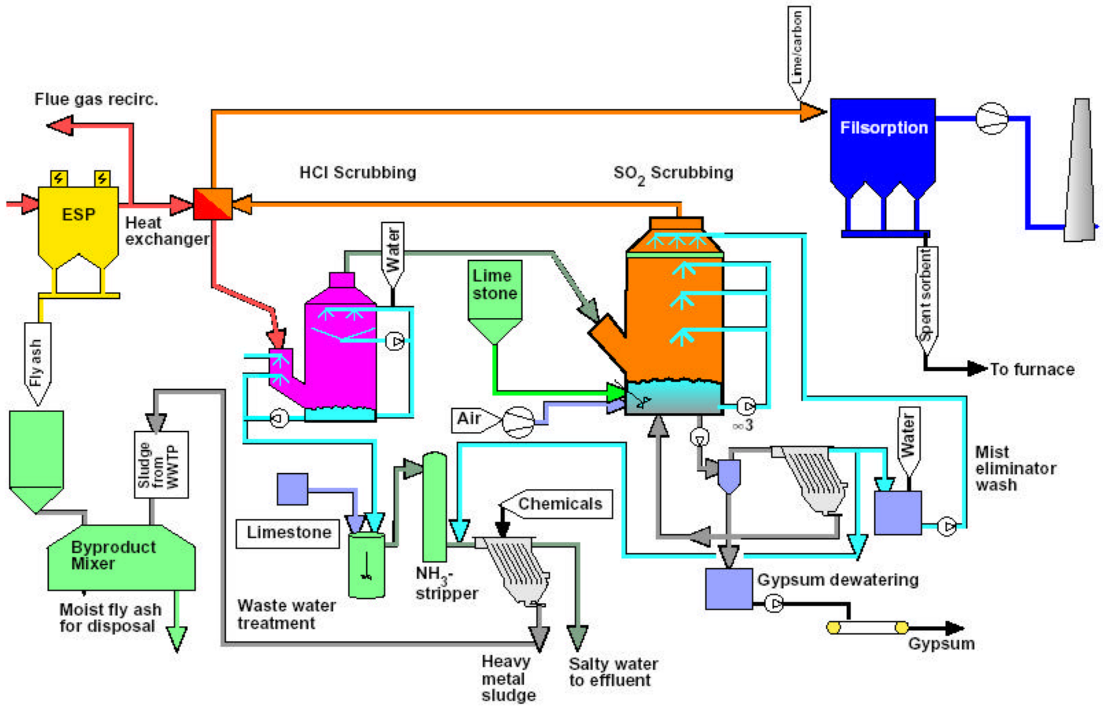


Fig 1. Typical gas cleaning system for several larger European WtE plants. These plants typically have flue gas flows of 100 000 acfm.

The gas cleaning system consists typically of ESP, acid scrubber where HCl is removed, SO2 scrubber (which can be of the limestone type) flue gas reheat and a secondary fabric filter, sometimes referred to as a police filter. The underlying driving forces for the design of these systems are minimization of end products and to achieve very low stack emissions.

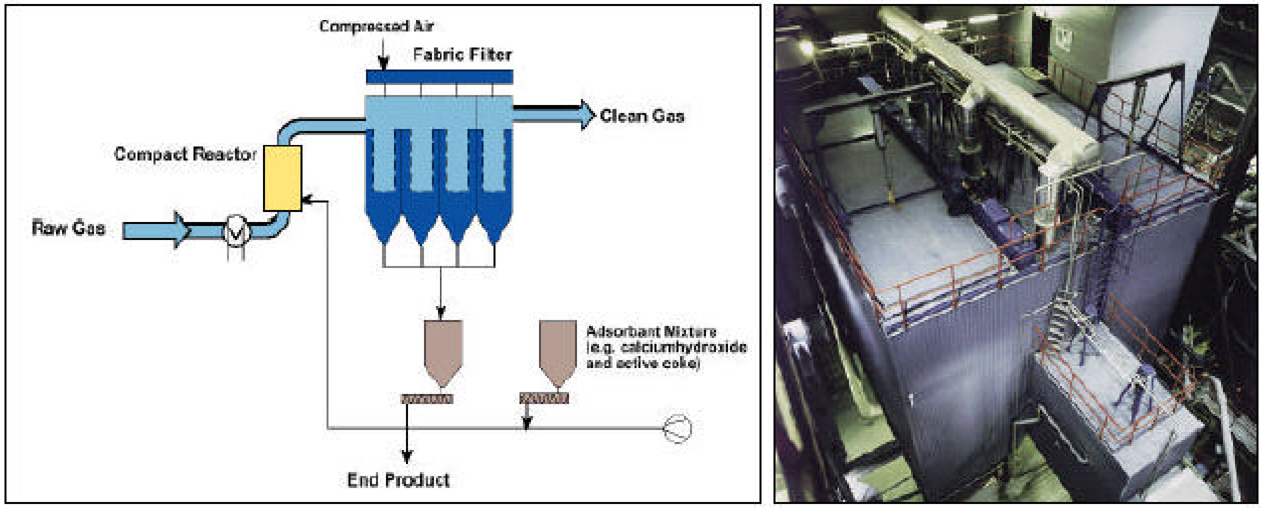


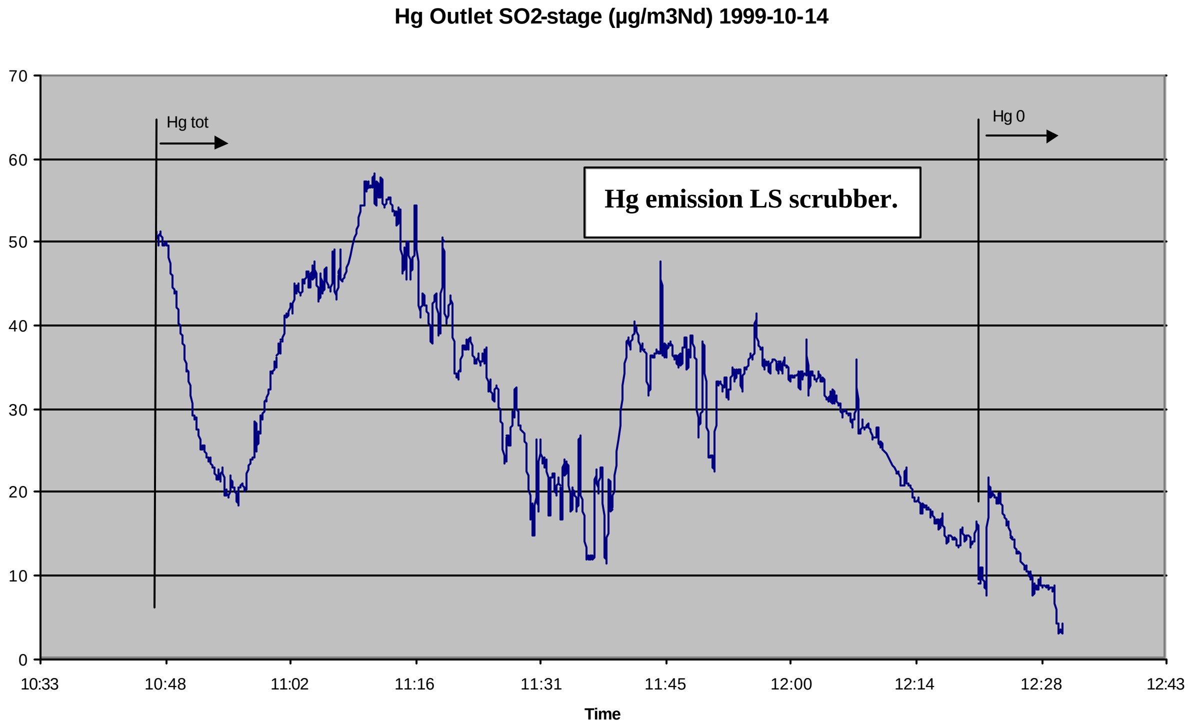
Fig 2. The 1992 Filsorption installation in Uppsala.

This PAC injection/fabric filter technology was pioneered by ALSTOM and the first system to be built was at the Uppsala incinerator in Sweden, which went into operation in 1992, and has been in continuous service since then. The unit serves a gas-flow of 120,000 scfm from three incinerator lines. Since then about two dozens of gas cleaning lines using PAC injection in filters have been built in Europe. Totally, ALSTOM built 18 plants of this type in the 90's having a total gas flow of 1100 acfm.

The flue gas composition after the various gas-cleaning steps is given in Table 1, and one can note that comparing to the task for the PAC-filter at a coal-fired boiler, the conditions differs mainly with respect to the flue gas SO_2 content which is a factor 10-100 times less than what is the case for US coal-fired plants. However, in the filter proper, both HCl and Hg are of the same order of magnitude as in coal firing. Another difference is that in WtE, the flue gas composition is more variable with time, due to a more varying fuel, but for the filter proper, the variations are to some extent evened out by the buffering capacity of the upstream scrubbers.

	After ESP	After acid Scrubber	After LS scrubber	After PAC filter
SO2 ppm	150	150	20	5
HCl ppm	900	50	5	1
Hg, mg/nm^3	150	50	35	3
Temp, ^{o}F	300	130	130	270

Table 1 Typical (average) flue gas compositions in the WtE gas cleaning train of Fig.1. based on measurements in the two sister plants KARA and Vestforbraending.



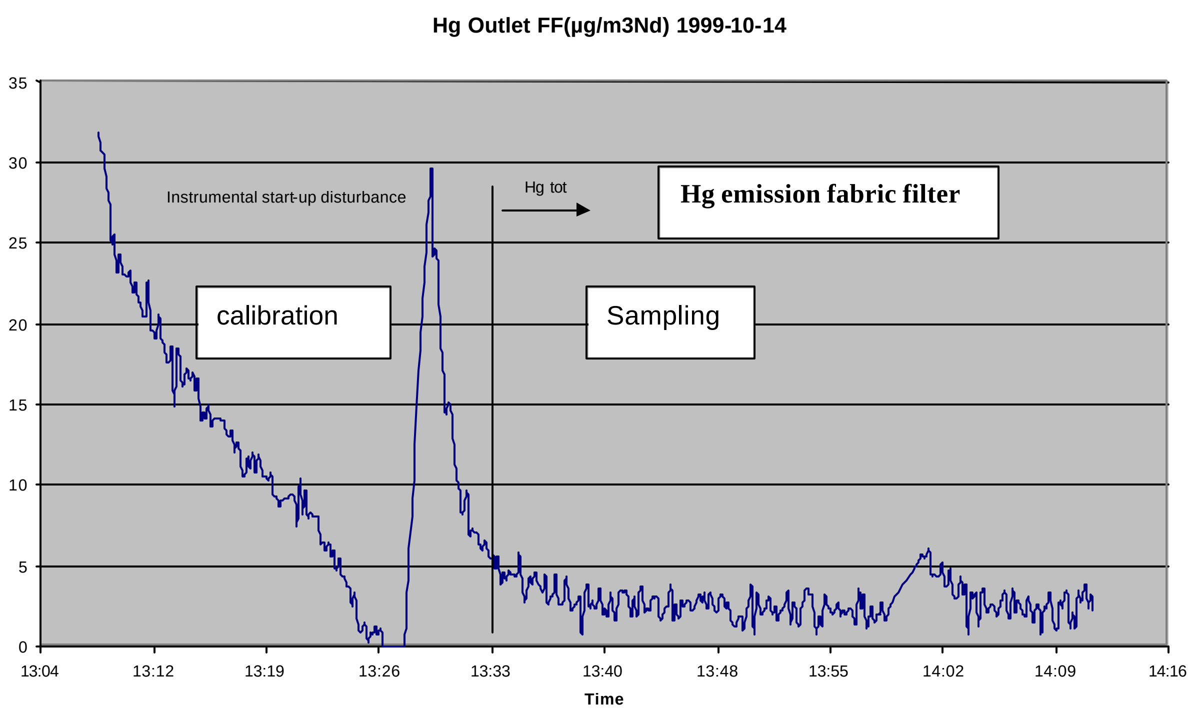


Fig 3. Flue gas total mercury at LS scrubber outlet/PAC filter inlet (top) and at PAC filter outlet (bottom). The high figures to the right in the bottom diagram reflect instrument tuning.

Dynamic mercury measurements in different locations at the Vestfoerbraending plant in Denmark performed by ALSTOM using Semtech UV spectrometry (and SnCl2 converter for speciation) for mercury determination is shown in the below Figure 3. We note that although the outlet mercury from the LS scrubber has a very pronounced high short-term variability, the filter mercury emission is almost constant, which reflects the buffering capacity of the PAC inventory on the bags. Time-average collection efficiency of mercury is 90 %. The PAC feeding rate is 1.5 lbs PAC/Macfm, and PAC feed is simply controlled on flue gas flow.

These filters have been equipped with PPS bags and with moderate filter velocities of 4 fpm giving very low pulsing frequency and mechanical wear, and since flue gas temperature is low and the flue gas is clean, the chemical degradation is very slow, which has resulted in extremely long bag life - the Uppsala bags have reportedly been in service for more than 10 years. Several of the plants having come on line later still operates with the original set of bags. Bag cleaning frequency is correspondingly low, 0.25-0.5 pulses/bag/hr. At the higher operational temperatures prevailing at coal-fired plants, the chemical degradation of the bag material will be more rapid and bag life will be shorter. Under such conditions, a more optimal solution is to design for a higher A/C ratio, balancing the mechanical and the chemical degradation, and as a result, unfortunately, a shorter bag life.

The filters on the dozen and a half ALSTOM plants in operation are of the LKP type, Fig 4.

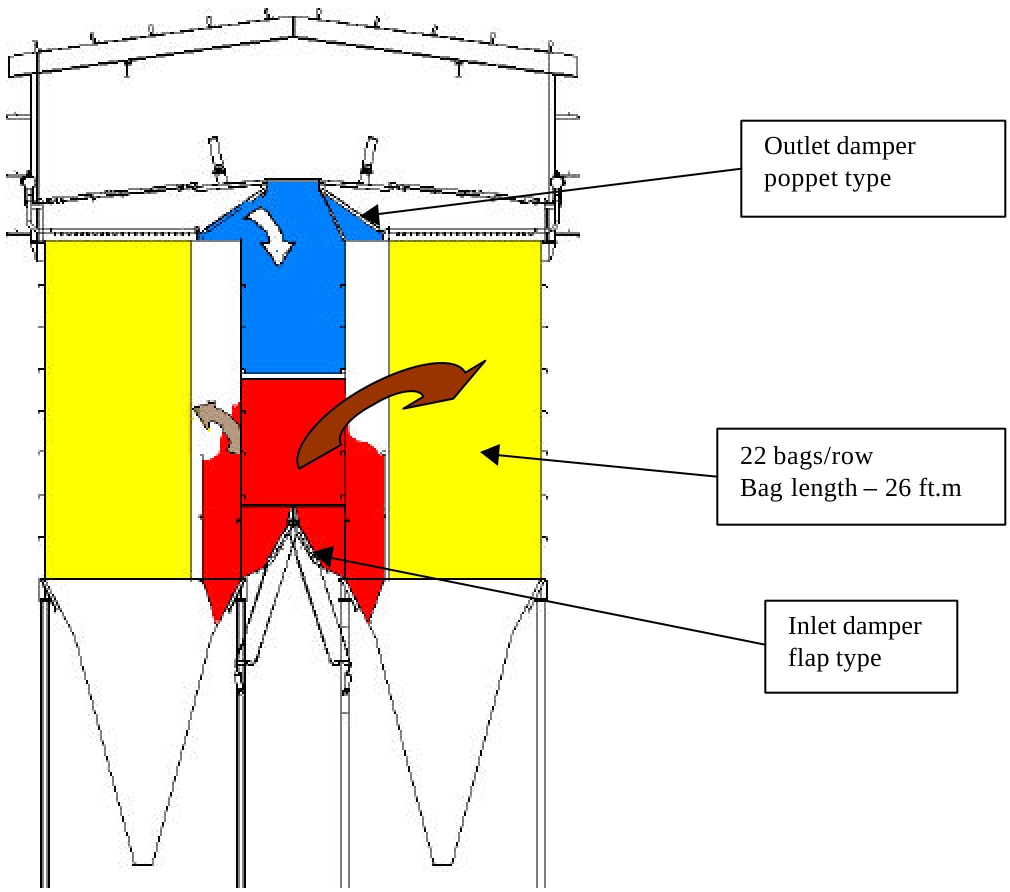


Fig. 4 The ALSTOM LKP type of bag filter employed for PAC injection for mercury control.

SORBENT HANDLING AND EXPERIENCES

The sorbent material in these plants consists of a mixture of commercially available PAC (20-50%) and calcium hydrate or finely ground limestone. The first reason for this admixture is that the alkali serves as additional acid gas absorbent, further decreasing the SO_2 and HCl emissions form the plant. For coal-fired power plants having a need for additional SO_3 control, that end can be achieved in the same way.

The other reason for admixture of inert alkali material is related to the risk for ignition during operational upset conditions. PAC is an oxidizable material, which under certain conditions is prone to autoignition. The mechanism derives from the fact that the material is slowly oxidizing with an accompanying heat production. Under most conditions, this oxidation is so slow, that it has no practical consequences. The resulting heat is dissipated to the environment via conduction through the bulk material and radiation and conduction at the boundaries. The oxidation rate is very dependant on temperature as well as on oxygen content. If the system is stable (heat production being effectively balanced by the heat dissipation, no appreciable temperature increase) or instable is given by the factors, time, temperature, oxygen content and material volume. A critical case may develop when a large volume of PAC is stored in a heated hopper for a long time in a high oxygen atmosphere. The conditions for autoignition can be quantitatively evaluated for different materials using standardized procedures, and dilution with inert material (flyash, alkali) will shift the autoignition criterion to more extreme conditions.

To build an inherently safe system taking all possible (including less probable) operational cases out of the envelope for possible autoignition with a generous margin, it appears good engineering practice to design the system meeting the following criteria:

- ash extraction from the filter needs to be continuous and reliably designed with minimal air in-leakage
- hopper valley angles need to be steep enough to ascertain fail-safe discharge during all operational conditions
- hopper heating needs to be uniform and controlled to prevent both condensation and local over-temperatures.
- the used product ash needs to be cooled before being discharged into the large volume storage silo
- compliance with the NFPA codes (notable codes 68, 650, 654, and 850).

CONCLUSIONS

The overall experience from the WtE field of PAC injection for mercury control is that systems, which have been designed following the considerations above, have given long and reliable service contributing to very low emissions from the plants so equipped. Since the gas conditions and the duty is rather close to that prevailing for mercury control on large coal-fired boilers, this experience is relevant and adds considerably to the confidence in the concept of PAC injection for mercury control on coal-fired boilers.

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